

MICROCOPY RESOLUTION TEST CHART



AD-A148 277

DTIC ELECTE NOV 2 1 1984

Approved for public released
Distribution Unlimited

84 11 16 020

# Investigations of Vacuum Ultraviolet and Soft X-ray Lasers

Report for
Grant period 1983-1984

Prepared by
Ahmet Elçi

Principle Investigators M. O. Scully and A. Elçi

Institute for Modern Optics

Department of Physics and Astronomy

University of New Mexico

Albuquerque, New Mexico 98131



B

DISTRIBUTION STATEMENT A

211

Approved for public release Distribution Unlimited

REPORT DOCUMENTATION PAGE	READ INSTRUCTIONS BEFORE COMPLETING FORM	
1. REPORT NUMBER 2. GOVT ACCESSION NO.	RECIPIENT'S CATALOG NUMBER	
AFOSR-TR- 11 0993 AD- A 148	\d'/'/	
4. TITLE (and Subtitle)	5. Type of REPORT & PERIOD COVERED Final Report 5/15/83-	
Investigations of Vacuum Ultraviolet and Soft X-ray Lasers	8/14/84	
bolt A-lay basers	6. PERFORMING ORG. REPORT NUMBER	
7. AUTHOR(*)	8. CONTRACT OR GRANT NUMBER(*)	
A. Elçi	AFOSR-81-0128 🏚	
9. PERFORMING ORGANIZATION NAME AND ADDRESS	10. PROGRAM ELEMENT, PROJECT, TASK AREA & WORK UNIT NUMBERS	
Institute for Modern Optics	6/102F	
Dep. Of Physics and Astronomy	2301/AS	
Univ. of New Mexico. Albuquerque, NM 87131	12. REPORT DATE	
Airforce Office of Scientific Research (AFOSR)	August 1984	
Bolling AFB, DC 20332	13. NUMBER OF PAGES	
14. MONITORING AGENCY NAME & ADDRESS(II different from Controlling Office)	15. SECURITY CLASS. (of this report)	
	Unclassified	
	15a, DECLASSIFICATION/DOWNGRADING SCHEDULE	
Approved for the did release; distribution in the different fro  17. DISTRIBUTION STATEMENT (of the abetract entered in Block 20, if different fro	- Recently	
18. SUPPLEMENTARY NOTES		
	·	
19. KEY WORDS (Continue on reverse side if necessary and identify by block number)	j	
X-ray Lasers, Surface Scattering, Free Electron Nuclear Spectroscopy	Laser,	
· (	ł	
20 <sub>c.\</sub> ABSTRACT (Continue on reverse side if necessary and identify by block number)		
We present the results of our investigations conscattered ions for x-ray lasers, constraints over free electron laser in the x-ray regime, and efficient on nuclear transitions.	r the operation of	
	Ì	

## TABLE OF CONTENTS

ï

		Page
I. INT	RODUCTION	1
II. SUR	REACE SCATTERING OF IONS AND SOFT X-RAY EMISSION	2
III. FRE	E ELECTRON LASER AT X-RAY WAVELENGTHS	6
IV. NUC	ILEAR SPECTROSCOPY	7
APPENDIX	A: "Stimulated Emission From Surface Scattered Ions"	
APPENDIX	B: "SOFT X-Ray Amplification by Ions Scattering From	
	Antiferromagnetic Surfaces"	
APPENDIX	C: "Magnetic Surface-Induced Spin-Flip Transitions in	
	Singlet 0 <sub>2</sub> "	
APPENDIX	D: "Free-Electron Lasers in the X-Ray Region"	
APPENDIX	E: "Forbidden Nuclear Beta-Decay in an Intense Plane	
	Wave Field"	
APPENDIX	F: "Comment on Enhancement of Forbidden Nuclear Beta	
	Decay by High-Intensity Radio-Frequency Fields"	

Accession	n For	- ( st )
NTIS GR DTI TIS Upon a con	i	1
10 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1	in / The Theat	
A 1	e e e e e e e e e e e e e e e e e e e	
A·1		

#### I. INTRODUCTION

During the grant period 5/15/83 - 8/14/84, the efforts of our group were directed towards investigating surface scattering of ions for the purpose of obtaining stimulated emission of soft x-rays, as well as scaling of free electron laser to x-ray wavelengths, and nuclear spectroscopy in the x-ray regime.

The following report consists of three sections and six appendices. The first section summarizes scattering of ions from different types of surfaces and stimulated emission of light from scattered ions. The second section summarizes our continuing work in the area of an x-ray free electron laser. Another continuing effort of our group has been in nuclear spectroscopy with x-ray lasers and intense fields. The work in this area, corresponding to the relevant grant period, is summarized in the third section. The papers which detail the results of these investigations either in published or preprint form are reproduced in the appendices.

#### II. SURFACE SCATTERING OF IONS AND SOFT X-RAY EMISSION

Coherent excitation of ions by the periodic potential of a crystal proposed by Okorokov sometime ago arises from the fact that an ion moving in a crystal with speed v experiences a time dependent potential with harmonics  $\Omega_n = (2\pi vn)/d$ , where n is a positive integer and d is the lattice spacing in the direction of ionic motion. If v and d are such that  $\Omega_n$  matches an ionic transition frequency, then ions are resonantly excited by the nth harmonic. probability that ions are in an excited state becomes a periodic function of the thickness of the foil, which can be viewed as a consequence of the Rabi oscillations of ions in their rest frame. One can expect the same physical picture to hold for ions scattering from crystal surfaces at grazing incidences. This may be called the surface-Okorokov effect and offers interesting possibilities for obtaining stimulated emission of soft x-rays from ions. One can coherently excite a beam of ions into certain high lying states by an adjustment of the beam velocity to the crystal periodicity, and obtain an effective population inversion relative to some low lying states. For such a purpose, the grazing angle surface scattering geometry has considerable advantages over the beam foil geometry in that the damage to the surface resulting from ion bombardment is minimized at grazing incidences and the same surface can be used repeatedly. Furthermore, stopping powers of surfaces can be lower than stopping powers of bulk materials, which means that ions can resonantly interact with surfaces for longer distances than with foils.

We analyzed the scattering of hydrogenic ions, such as  ${\rm Li}^{+2}$ ,  ${\rm Be}^{+3}$ , etc., from crystal surfaces at grazing incidences, and showed that ions execute extremely rapid Rabi oscillations in their rest frame ( ${\sim}10^{17}~{\rm sec}^{-1}$ ). We assumed that the ionic beam simultaneously interacts with a coherent electromagnetic field whose frequency matches one of the ionic transition frequencies

corresponding to an effective population inversion, and showed that the inhomogeneously broadened gain of the signal can be substantial, on the order of 1 cm<sup>-1</sup>, for reasonable beam currents such as 4.8 ampere/mm<sup>2</sup>. The analysis is detailed in the preprint in Appendix A, which is accepted for publication in Phys.Rev. Letters.

In the free electron laser, lasing occurs at the wavelength  $\lambda \simeq 2\lambda_{\rm q}/\gamma$ , where  $\lambda_{\rm q}$  is the wiggler wavelength of the magnetic selonoid and  $\gamma$  is the ratio of the electron's energy to its rest mass energy. In principle one can decrease  $\lambda$  either by increasing the electronic energy or by decreasing the wiggler wavelength  $\lambda_{\rm q}$ . The latter method is not very practical with mechanical systems used in free electron lasers. There are, however, systems which are periodic on atomic scale, and which might be used as short wavelength wigglers for electrons or ions. An interesting idea is to use antiferromagnetic crystals as wigglers for ions. These crystals have strong internal magnetic fields that are periodic with some lattice constant and can be used to excite ions coherently. Excited ions can then decay radiatively and thus convert the virtual photons of the material magnetic field to real photons.

We analyzed the problem for a beam of hydrogenic Be<sup>+3</sup> ions scattering from the surface of an antiferromagnetic material such as  $FeF_2$  or MnO. In the geometry we considered, Be<sup>+3</sup> ions penetrate just below the surface of the material, travel some distance L, and then come out. They simultaneously interact with soft X-Ray signal of frequency  $\hbar\omega_S \sim 30$  eV which matches the frequency  $\omega_{32}$  of the n=3  $\rightarrow$  n=2 transitions of Be<sup>+3</sup>. We first considered the magnetization fields inside an antiferromagnet and deduced the general form of the fields from a simple argument. We then obtained a simple set of equations of motion for the ionic amplitude from the Schrodinger equation in the rest frame of an ion. These equations show that ions in their rest frame execute Rabi oscillations between the states n = 1 and n = 3, if the ion velocity is

properly adjusted. The Rabi frequency  $\Omega_{\rm R}$  is proportional to B<sub>o</sub>, the amplitude of the magnetization of one of the sublattices of the antiferromagnet, and to v, the ion velocity.  $\Omega_{R}$  is on the order of  $4x10^{13}~\text{sec}^{-1}$  for v =  $10^{9}~\text{cm/sec,}$  which is nearly four orders of magnitude larger than the spontaneous radiative decay rate of the n = 3 states of  $Be^{+3}$ . Because ions are actually moving with velocity v, the Rabi oscillation in the rest frame become spatial oscillations in the lab frame. If  $L_p$  designates the spatial interval for half a Rabi cycle (i.e.,  $L_{\rm R}$  is the minimum distance required for an ion to travel in the material in order to get the ion fully excited into the n = 3 states, if it is initially in the ground state), then  $L_{\rm R}$  is on the order of  $10^{-4}$  cm. We also analyzed the gain of the signal field in the weak signal regime. Due to the coherent excitation of ions, the expression for the gain differs from the usual small-signal swept-gain, and have a number of novel features. It turns out that only for a narrow region of detuning between  $\omega_{\rm e}$  and  $\omega_{3,2}$  can one get finite gain. The maximum gain is about  $10~\mathrm{cm}^{-1}$  if the ionic density is on the order of  $10^{1}$ cm<sup>-3</sup>. Appendix B reproduces a preprint which gives the details of these investigations.

Our work with magnetic surfaces has yielded another idea which might lead to a new class of lasers. In diatomic molecules formed by the group VI elements (e.g.,  $O_2$ ,  $S_2$ ,  $S_3$ ) and by a combination of group V and group VII elements (e.g., NF, NC1, PC1), the spin selection rule  $\Delta S = 0$  leads to highly metatable states which can be very useful for high power laser systems. A well-known case is the  $\frac{1}{\Delta}_g$  state of the oxygen molecule. Due to spin and parity symmetry, its decay to the ground state  $\frac{3}{\Delta}_g^{-1}$  is highly forbidden with spontaneous decay rate 2.6 x  $10^{-6}$  sec<sup>-1</sup>. This fact is used in oxygen-iodine lasers to store the energy in the  $\frac{1}{\Delta}_g$  state of  $O_2$ , which is then collisionally transferred to atomic iodine via the reaction  $O_2$  ( $\frac{1}{\Delta}_g$ ) +  $\frac{1}{\Delta}_g$  +  $\frac{1}$ 

place on the iodine through its decay back to  ${}^{2}P_{1/2}$ , which is a magnetic-dipole allowed transition with a spontaneous transition rate of 7.7 sec-1. The frequencies of  ${}^{1}\Delta_{g}$  -  ${}^{3}\Sigma_{g}^{-}$  and  ${}^{2}P_{1/2}$  -  ${}^{2}P_{3/2}$  transitions match, which makes it possible to resonantly transfer energy between O2 and I. In practice it is difficult to find such matching partners for other metastable systems which might have applications to high power lasers. For this reason, as well as to avoid loses involved in collisional energy transfers, there has long been an interet in obtaining radiative emissions directly from the excited metastable system itself. To achieve direct radiative emission, it is necessary to break the various symmetries of the metastable system. For instance, the parity symmetry can be broken by applying static electric field or nonresonant radiation, or by means of nonresonant collisons with another molecular or atomic system. On the other hand, breaking of the spin symmetry requires application of either a spatially varying magnetic field as in a Stern-Gerlach device, or a direct spin-spin coupling another system. This latter possibility can be realized by putting the singlet delta oxygen in contact with a magnetic surface, where the electrons of the singlet delta oxygen will couple to the external spins through their internal magnetic moments. By means of a simple perturbation analysis, we showed that both parity and spin symmetries are broken and that an electric dipole moment is induced between  $^1\Delta_{\sigma}$  and  $^3\Sigma_{\sigma}^-$  which is on the order of  $10^{-2}$  Debye. This implies a spontaneous transition rate of about 102 sec-1, which is nearly six orders of magnitude larger than the corresponding rate for isolated  $O_2$ . The analysis should also hold for NF which has an ultraviolet transition frequency between the electronic ground states and the first excited singlet delta states. This work is already published (Phys. Rev. A 29, February, 1984). The paper is reproduced in Appendix C.

## III. FREE ELECTRON LASER AT X-RAY WAVELENGTHS

An x-ray free electron laser must operate with large gain per pass because of the poor quality of available mirrors at x-ray frequencies. Moreover, the effects of inhomogeneous broadening, start-up from noise, coherence development, and quantum recoil can be important in determining whether and how such devices will operate. We performed one-dimensional classical and semiclassical analyses of the gain regimes for x-ray free electron lasers. Based on these analyses, we determined some of the constraints on such devices imposed by effects such as diffractive spreading of the laser beam, energy spread and emittance of the electron beam, and transverse variations in the wiggler field. These constraints are discussed in the preprint reproduced in Appendix D.

#### IV. NUCLEAR SPECTROSCOPY

In two recent papers [Phys. Rev. C  $\underline{27}$  (1984), 1229], Reiss claimed that forbidden nuclear beta decay can occur in the presence of an intense but low frequency electromagnetic field. His approach treats the weak coupling to first order, and the electromagnetic field as nearly as possible to all orders. Thus, he describes electron states by Volkov wavefunctions, whereas he approximates nuclear states by the so-called momentum-translation-approximation wavefunctions. We have now shown by explicit calculation that within this model, the total beta decay is essentially independent of the external 'electromagnetic field. This conclusion holds both in nonrelativistic and relativistic treatments of the problem. Moreover, by comparing the " $\mathbf{r}$  ·  $\mathbf{E}$ " and " $\mathbf{p}$  ·  $\mathbf{A}$ " form of the interaction Hamiltonian, we have shown that the waveful continuous momentum translation approximation is the unperturbed state in the Coulomb gauge. This work is reported in a preprint and a paper which appeared in Phys. Rev. C 29, March 1984. These are reproduced in Appendices E and F, respectively.

## APPENDIX A

STIMULATED EMISSION FROM SURFACE SCATTERED IONS

(accepted for publication in Phys. Rev. Lett.)

Ahmet Elçi

Institute for Modern Optics

Department of Physics and Astronomy

University of New Mexico

Albuquerque, New Mexico 87131

### Abstract

We discuss coherent excitation of ions incident on crystal surfaces at grazing angles and show that substantial gains for soft x-rays can be achieved for reasonable beam currents.

Coherent excitation of ions by the periodic potential of a crystal was proposed by Okorokov sometime ago [1] and has been observed by different groups who passed highly ionized atoms through thin foils [2,3]. The effect arises from the fact that an ion moving in a crystal with speed v experiences a time dependent potential with harmonics  $\Omega_n = (2\pi vn)/d$ , where n is a positive integer and d is the lattice spacing in the direction of ionic motion. If v and d are such that  $\Omega_n$  matches an ionic transition frequency, then ions are resonantly excited by the  $n^{\frac{th}{n}}$  harmonic. The probability that ions are in an excited state becomes a periodic function of the thickness of the foil [4], which can be viewed as a consequence of the Rabi oscillations [5] of ions in their rest frame. When one goes to the laboratory frame in which the crystal is at rest, the Rabi oscillations in time become thickness dependent oscillations in space. The Okorokov effect depends critically on the finiteness of the probability that ions keep their charge states constant without further ionization or neutralization in the crystal.

One can expect the same physical picture to hold for ions scattering from crystal surfaces at grazing incidences. This may be called the surface-Okorokov effect and offers interesting possibilities for obtaining stimulated emission of soft x-rays from ions. One can coherently excite a beam of ions into certain high lying states by an adjustment of the beam velocity to the crystal periodicity, and obtain an effective population inversion relative to some low lying states. For such a purpose, the grazing angle surface scattering geometry has considerable advantages over the beam foil geometry. The damage to the surface resulting from ion bombardment is minimized at grazing incidences so that the same surface can be used repeatedly [6]. Furthermore, stopping powers of surfaces can be lower than stopping powers of bulk materials, which means that ions can resonantly interact with surfaces for longer distances than with foils.

In this paper we consider the scattering of hydrogenic ions, such as Li<sup>+2</sup>, Be<sup>+3</sup>, etc., from crystal surfaces at grazing incidences, and show that ions execute extremely rapid Rabi oscillations in their rest frame. We assume that the ionic beam simultaneously interacts ith a coherent electromagnetic field whose frequency matches one of the ionic transition frequencies corresponding to an effective population inversion, and show that the inhomogeneously broadened gain of the signal can be substantial.

The geometry of the problem is shown in Fig. 1. An ion is incident from the left, moving in the z'-direction (primed coordinates refer to the laboratory frame, unprimed ones to the rest frame of the ion). In the laboratory frame, the periodic crystal potential can be described by

$$V_{\mathbf{c}}'(\vec{x}') = \theta(x_0 - x') \sum_{\vec{G}} q_{\vec{G}} e^{i\vec{G} \cdot \vec{x}'}$$
(1)

Here,  $x_0$  designates the distance between the c.o.m. of the ion and the surface  $(x_0)$  is negative if the ion is above the surface;  $x_0$  is positive if the ion penetrates the surface, and is actually inside the crystal).  $\vec{G}$ 's are the reciprocal lattice vectors. We have assumed that the crystal terminates sharply on a plane at  $x_0$ . This plane need not coincide with the plane of the first layer of surface atoms; rather, it should be considered as an effective plane where the influence of surface atoms and electrons of the crystal on the ion begins to be appreciable.

If one goes to the ionic rest frame,  $V_c^*(\vec{x}^*)$  is transformed into a vector  $(\vec{A}_c)$  and a scalar potential  $(V_c)$ .  $\vec{A}_c$  is purely longitudinal, and can be eliminated by a gauge transformation on the wavefunction of the ionic electron,  $\psi(x,t) = \exp[i\Lambda(\vec{x},t)] \cdot \phi(\vec{x},t)$ , such that  $\nabla \Lambda = (e/\hbar c)\vec{A}_c$ . One finds that  $\phi$  satisfies the Schrödinger equation

$$i\hbar \partial_{t} \phi = [(2m)^{-1}(\vec{p} - e\vec{A}_{s}c^{-1})^{2} - Ze^{2}|\vec{x}|^{-1} + V_{\perp} + V_{\parallel}]\phi$$
 (2a)

where

$$V_{\perp}(\vec{x}_{\perp}) = e\gamma\theta(x_0 - x)\sum_{s\perp} w_{\vec{G}} e^{i\vec{G} \cdot \vec{x}}, \quad s\perp = \{\vec{G} | \hat{z} \cdot \vec{G} = 0\}, \qquad (2b)$$

$$V_{\|}(\vec{x},t) = e\gamma^{-1}\theta(x_0 - x)\sum_{s\|} v_{\vec{G}} e^{i\vec{G} \cdot \vec{x}}, \quad s\| = \{\vec{G} | \hat{z} \cdot \vec{G} \neq 0\},$$
 (2c)

and  $\vec{A}_s = y\vec{A}_s \cos(k_s z - \omega_s t)$  is the signal field.  $\gamma$  is the relativistic factor  $(1-v^2/c^2)^{-\frac{1}{2}}$  and essentially equals 1 for all v's of interest here.  $V_{\perp}$  is a static potential that depends only on the transverse coordinates of the ionic electron. Note that going from the laboratory frame to the ionic rest frame is equivalent to taking a time average of the crystal potential along the trajectory of the ion;  $V_i$  is this time averaged potential.  $V_i$  is obtained from this perspective in Refs. 7 and 8, and the level shifts induced by  $\mathbf{V}_{\underline{\mathbf{I}}}$  on ionic spectra in foils are estimated in Ref. 8. Here, however, we will ignore  $\mathbf{V}_{\!\!1}$  , assuming that it produces negligible perturbations. For our discussion, the important term is the time dependent potential  $\boldsymbol{V}_{\parallel}$  . If the basis vectors of the reciprocal lattice space are  $\vec{G}_1$ ,  $\vec{G}_2$  and  $\vec{G}_3$ , that is  $\vec{G}$  =  $\ell \vec{G}_1$  +  $m\vec{G}_2$  +  $n\vec{G}_3$ , where  $\ell,m,n$  are integers, then the harmonics of  $V_{||}$  are given by  $\gamma v\hat{z} \cdot (z\vec{G}_1 + m\vec{G}_2 + n\vec{G}_3)$ . In the following, we will consider the case in which one of the fundamental frequencies obtained from this expression equals or is near an ionic transition frequency. Specifically, we will assume that only n = 1 and n = 3 levels of the ion are involved in the transitions induced by the crystal, and that an effective population inversion occurs between n = 3 and

n = 2 levels as shown in Fig. 1. One can then ignore all  $\vec{G}$ 's in the set s|l except for those which give a constant frequency  $\Omega$  which is either resonant or nearly so with the ionic transition frequency  $\omega_{31}$ . Thus we define the set of vectors  $\vec{g}$  such that  $\vec{g} = \hat{x}G_X + \hat{y}G_Y + \hat{z}\gamma G_Z$  and  $|\vec{g}\cdot\hat{z}| = \text{constant} = \Omega/v$ . Ignoring the signal field for the moment, the amplitude equations in the rotating wave approximation are

$$i\hat{c}_{100} = e^{i(\Omega - \omega_{31})t}$$
 $i\hat{c}_{100;3lm} c_{3lm}$ , (3a)

$$i\dot{c}_{3\ell m} = e^{-i(\Omega - \omega_{31})t} \star M_{100;3\ell m} c_{100},$$
 (3b)

where

$$M_{n\ell m; n'\ell'm'} = e(\hbar\gamma)^{-1} \sum_{\overrightarrow{g}} w_{\overrightarrow{g}} \int d\overrightarrow{x} \ u_{n\ell m}^{*}(\overrightarrow{x}) \theta(x_{0} - x) e^{i\overrightarrow{g} \cdot \overrightarrow{x}} u_{n'\ell'm'}(\overrightarrow{x}) , \qquad (3c)$$

and  $u_{nlm}$ 's are the hydrogenic wavefunctions. Assuming  $c_{100}(0) = 1$  and  $c_{3lm}(0) = 0$ , the solution of (3a,b) is

$$c_{100}(t) = \left(\Omega^{+} e^{-i\Omega^{-}t} - \Omega^{-} e^{-i\Omega^{+}t}\right) / (\Omega^{+} - \Omega^{-}) ,$$
 (4a)

$$c_{3lm}(t) = M_{100;3lm}^{\star} \left( e^{i\Omega^{\dagger}t} - e^{i\Omega^{-}t} \right) / (\Omega^{\dagger} - \Omega^{-}) ,$$
 (4b)

where  $\Omega^{\pm}(\ell m) = (\omega_{31} - \Omega)/2 \pm [(\omega_{31} - \Omega)^2/4 + \Omega_{R\ell m}^2]^{\frac{1}{2}}$  and  $\Omega_{R\ell m} = [M_{100;3\ell m}]$  is the Rabi frequency.

If the ion penetrates the surface and  $|\mathbf{x}_0| \sim l \text{Å} > (a_B/Z)$ , one can essentially ignore the step function in (3c) since  $u_{n \ell m}$ 's are localized to a radius of approximately  $(na_R)/Z$ . One then finds

$$M_{100;300}^{(p)} = 3\sqrt{3\pi} e(4\hbar\gamma)^{-1} \sum_{g} \psi_{g}^{4} (\beta_{g}^{2} + 3) (\beta_{g}^{2} + 1)^{-4} , \qquad (5a)$$

$$M_{100;31m}^{(p)} = i\sqrt{\pi} e(2\hbar\gamma\sqrt{3})^{-1} \sum_{g} \varphi_{g}^{3} (\beta_{g}^{2} + 2)(\beta_{g}^{2} + 1)^{-3} Y_{1m}(\hat{g}) , \qquad (5b)$$

$$M_{100;51m}^{(p)} = \frac{9e^{2}}{2\hbar\gamma} \sqrt{\frac{3}{\pi}} \sum_{g} v_{2m} (\hat{g}) \beta_{g}^{3}$$

$$\times \left\{ \frac{\beta_{g} (3\beta_{g}^{3} + 4)}{6(\beta_{g}^{2} + 1)^{4}} - \sqrt{\frac{2}{\pi}} \sum_{s=0}^{\infty} \frac{\Gamma\left(s + \frac{5}{2}\right) \Gamma\left(s - \frac{3}{2}\right)}{2^{s} \Gamma\left(s + 1\right) \Gamma\left(s + \frac{7}{2}\right)} \left(1 - \frac{\beta_{g}}{\sqrt{\beta_{g}^{2} + 1}}\right)^{s + \frac{5}{2}} \right\}, (5e)$$

where  $\beta_g = 4Z/(3ga_B)$  and the superscript p is meant to imply a penetrating ion. Note that only  $M_{100;310}^{(p)}$  describes an electric dipole transition; the other transitions violate the dipole selection rules since they arise from higher order multiple transitions. This is in contrast to the formulation of the Okorokov effect in Rfs. 4 and 9, where the dipole approximation is assumed to hold. In justification of (5a-c), we note that whether one can make the dipole approximation depends on the ratio of the crystal period to the radius of the ion's electronic orbital, that is on whether  $|\vec{g}|(a_B^n/Z) \ll 1$ . For typical crystals one can easily obtain  $|\vec{g}|(a_B^n/Z) \ll 1$  even for low lying orbitals. The dipole approximation breaks down. Incidentally, this fact can be exploited to populate those states which are not dipole connected to the ground state and therefore, relatively long-lived. After leaving the surface, the ion remains in

a superposition of these long-lived excited states with the ground state. This superposition is stable relative to radiative decays to the ground state.

If one takes the pseudopotential values of  $\psi$  that are used in structure calculations [10], and uses just the smallest and the next smallest  $\vec{g}$ 's in the sums of (5), one finds for a crystal like Si that  $|M_{100;300}^{(p)}|$ ,  $|M_{100;32m}^{(p)}| \sim 10^{16}$  sec<sup>-1</sup> and  $|M_{100;310}^{(p)}| \sim 10^{17}$  sec<sup>-1</sup>. The resonance condition  $v\hat{z} \cdot (\vec{G}_1 + \vec{G}_2 + \vec{G}_3)$   $\omega_{31} \sim 193$  eV can be satisfied for a Be<sup>+3</sup> ion moving with speed  $v \sim 10^9$  cm/sec. It follows that the ion can be completely excited after traveling a "Rabi distance"  $\ell_{R2m} = \pi v/\Omega_{R\ell m} \sim 3 - 30 \text{Å}$ .

An interesting feature of (5b,c) is that the contributions of different components are proportional to  $Y_{lm}(\hat{g})$ . There are, therefore, orientation dependent interferences among  $\psi$  components. By varying the direction of the ionic beam, it is possible to pick different sets of  $\hat{g}$ -vectors, and thus obtain information about different  $\psi$  components from a comparison of Rabi frequencies corresponding to these sets.

We now consider the coupling of the ion to the coherent signal field and treat this coupling as a perturbation imposed upon the evolution described by (4a,b). In the calculation of the gain the steps are: a) determine  $c_{2\ell m}(t)$  to first order in  $A_s$  using  $c_{3\ell m}(t)$  given by (4b); b) calculate the transverse current density resulting from  $(3\ell m) + (2\ell ! n!)$  transitions; c) substitute this transverse current density into the Maxwell equation for  $A_s$  to obtain a dispersion relation between a real  $\omega_s$  and a complex  $k_s$ . The imaginary part of  $k_s$  gives the small signal gain for ions moving at a fixed velocity v. Finally, d) multiply the resulting gain with the probability distribution of ion velocities,  $\rho_{ion}(v)$ , and replace  $\Omega$  by  $v|\hat{z}\cdot\hat{g}|$ . We assume that the width of  $\rho_{ion}(v)$  is sufficiently narrow as not to effect the overall choice of the set of  $\hat{g}'s$ , and integrate over the interval  $-\infty < v < \infty$ . In the limit that the

lifetimes of the states in the n=2 shell go to  $\infty$ , the resulting inhomogeneously broadened gain is given by

$$g_{s} = \frac{2\pi^{2} \hbar e^{2} P_{cs} N_{ion}}{\omega_{s} m^{2} c} \sum_{\ell=0}^{2} \sum_{m=-\ell}^{+\ell} \frac{D_{\ell m} P_{ion} (v_{\ell m})}{[(\omega_{s} - \omega_{32})^{2} + \Omega_{R\ell m}^{2}]}$$
(6a)

where N is the density of ions, P is the probability that the charge state of ions remains unchanged, and  $v_{\ell m}$  is a tuning velocity given by

$$v_{\ell m} = \frac{1}{|\hat{z} \cdot \vec{g}|} \left( \omega_{s} - \omega_{32} - \frac{\Omega_{R\ell m}^{2}}{\omega_{s} - \omega_{32}} + \omega_{31} \right)$$
 (6b)

 $\mathbf{D}_{\text{0.m}}$  is a coefficient related to the momentum matrix elements,

$$D_{\ell m} = \sum_{\ell' m'} \sum_{\ell'' m''} \mathcal{P}_{2\ell' m'}; 3\ell'' m'' \mathcal{P}_{2\ell' m'}; 3\ell m M_{100; 3\ell m} M_{100; 3\ell'' m''}, \qquad (6c)$$

where

$$\begin{aligned} \mathbf{P}_{2\ell m;\,3\ell'm'} &= \int_{0}^{+\infty} \mathbf{u}_{2\ell m}^{*}(-i\nabla)\mathbf{u}_{3\ell'm'} \\ &= -(Z/a_{B})\delta_{m,0}(\delta_{m',1} + \delta_{m',-1})(\delta_{\ell,0}\delta_{\ell',1}\sqrt{n_{1}} + \delta_{\ell,1}\delta_{\ell',2}\sqrt{n_{2}}). \end{aligned} \tag{6d}$$

Here  $\eta_1=2^{13}\cdot 3^2\cdot 5^{-10}\simeq 7.6\mathrm{x}10^{-3}$  and  $\eta_2=2^{18}\cdot 3^3\cdot 5^{-10}\simeq 0.72$  are pure numbers [11]. A representative gain curve vs.  $\omega_s$  is depicted in Fig. 2 just for one particular (2m) in the sum (6a), and for a gaussian probability distribution of average velocity  $\mathbf{v}_0$  and width  $\Delta\mathbf{v}$ :  $\rho_{\mathrm{ion}}(\mathbf{v})=(\Delta\mathbf{v}\sqrt{\pi})^{-1}\exp[-(\mathbf{v}-\mathbf{v}_0)^2/\Delta\mathbf{v}^2]$ . As seen from the figure, the gain curve has double peaks surrounding the exact resonance at  $\omega_s=\omega_{32}$ , where it vanishes.  $\mathcal{G}_s$  is the sum of several such curves

which have differing heights and widths, but always vanish on exact resonance,  $\omega_s = \omega_{32}$ .

Another interesting feature of  $\mathcal{G}_{_{\rm S}}$  given by (6a) is that instead of the usual population inversion factor  $(N_{\rm upper} - N_{\rm lower})$ , it has an effective population inversion factor  $N_{\rm ion}\Omega_{\rm Rlm}^2/[(\omega_{_{\rm S}} - \omega_{_{32}})^2 + \Omega_{\rm Rlm}^2]$ . Thus all ions participate in the photon emission and absorption processes. The degree of this participation is determined by the coherence of ions and the corresponding Rabi frequency caused by the crystal.

Going back to the Be<sup>+3</sup> example, one finds (assuming P<sub>CS</sub>  $\sim$  .5)  $\mathcal{G}_{\rm S} \sim ({\rm N_{ion}}/10^{12}~{\rm cm^{-3}}) \times ({\rm v/\Delta v}) \times 10^{-4}~{\rm cm^{-1}} \ {\rm for\ a\ signal\ of\ }_{\omega_{\rm S}} \sim 30~{\rm eV}. \ {\rm Thus,\ for\ } \Delta {\rm v/v} \sim 10^{-4} \ {\rm and\ an\ ionic\ current\ density\ j_{\rm ion}} \sim 4.8~{\rm ampere/mm^2}, \ {\rm one\ has}$   $\mathcal{G}_{\rm c} \sim 1~{\rm cm^{-1}}.$ 

As an ion moves inside the crystal or on its surface, it loses energy to the material electrons, and its c.o.m. slows down. This can cause the ion to move away from the resonance with the crystal field. Scaling from the stopping powers of protons in bulk materials [12], one finds S  $\sim$  10 $^9$  eV/cm for Be $^{+3}$  with v  $\sim$  10 $^9$  cm/sec for a Si target. For grazing angle scatterings one can expect S to be lower since the number of material electrons may be depleted near the effective plane at  $x_0$  due to the formation of inversion layers further down in the interior [13]. It is also possible to choose the beam direction in such a way that ions can move between the crystal planes, avoiding heavy concentration of material electrons. Such a channeling of ions can decrease S by orders of magnitude [14]. For an ion moving the Rabi distance  $\ell_R$ , the minimum energy loss is  $E_{loss} \sim S\ell_R$ . Taking S  $\sim$  10 $^8$  eV/cm for channeled ions and  $\ell_R \sim 3$  – 30Å, one finds  $\Delta E_{loss} / E_{loss}$ 

It is a pleasure to acknowledge several useful conversations with Prof. M. O. Scully. This work received support from the Air Force Office of Scientific

Research and the Center for High Technology Materials of the University of New Nexico.

#### REFERENCES

- 1. V. V. Okorokov, JETP Lett. 2 (1965) 111.
- 2. V. V. Okorokov, et al., Phys. Lett. 43A (1973) 485.
- 3. S. Datz, et al., Phys. Rev. Lett. 40 (1978) 843.
- 4. J. Kondo, J. Phys. Soc. Japan 36 (1974) 1406.
- 5. P. L. Knight and P. W. Milloni, Physics Reports 66 (1980) 21.
- 6. C. Rau and R. Sizman, "Electron spin polarization at ferromagnetic single crystalline surfaces...", in <u>Atomic Collisions in Solids</u>, Vol. 1, eds. S. Datz, B. R. Appleton, and C. D. Moak (Plenum Press, New York 1975).
- 7. C. D. Moak, et al., Phys. Rev. <u>Al9</u> (1979) 977.
- 8. O. H. Crawford and R. H. Ritchie, Phys. Rev. A20 (1979) 1848.
- 9. S. Shindo and Y. H. Ohtsuki, Phys. Rev. B14 (1976) 3629.
- 10. V. Heine and D. Weaire, Solid State Physics, Vol. 24, pg. 358.
- 11. The signal-ion coupling is assumed to obey the dipole approximation since the signal wavelengths for soft x-rays are much larger than  $(na_R/Z)$ .
- 12. E. C. Montenegro, et al., Phys. Lett. 92A (1982) 195.
- 13. F. Stern and W. E. Howard, Phys. Rev. 163, 816 (1967).
- 14. D. S. Gemmell, Rev. Mod. Phys. 46 (1974) 129.

### FIGURE CAPTIONS

- FIG.1. The schematic of the grazing incidence surface scattering.
- FIG.2. The gain for a particular (£m) vs.  $\delta_S = (\omega_S \omega_{32}) (|\hat{z} \cdot \hat{g}| \pm v)^{-1}$ . For illustrative purposes,  $\delta_R = \Omega_R (|\hat{z} \cdot \hat{g}| \Delta v)^{-1}$  and  $\delta_C = (\omega_{31} v_0 |\hat{z} \cdot \hat{g}|) (|\hat{z} \cdot \hat{g}| \pm v)^{-1}$  are taken to be 3 and 1, respectively.

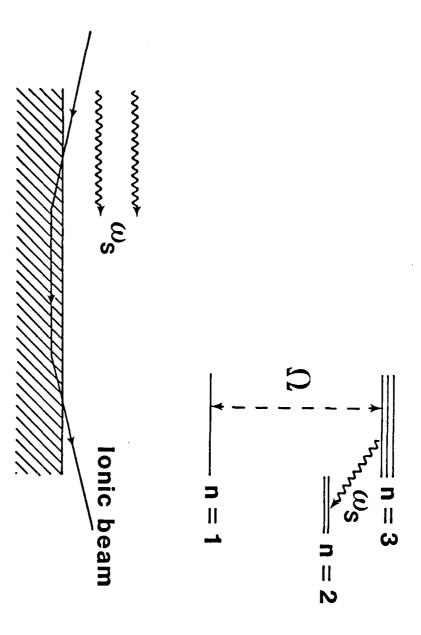
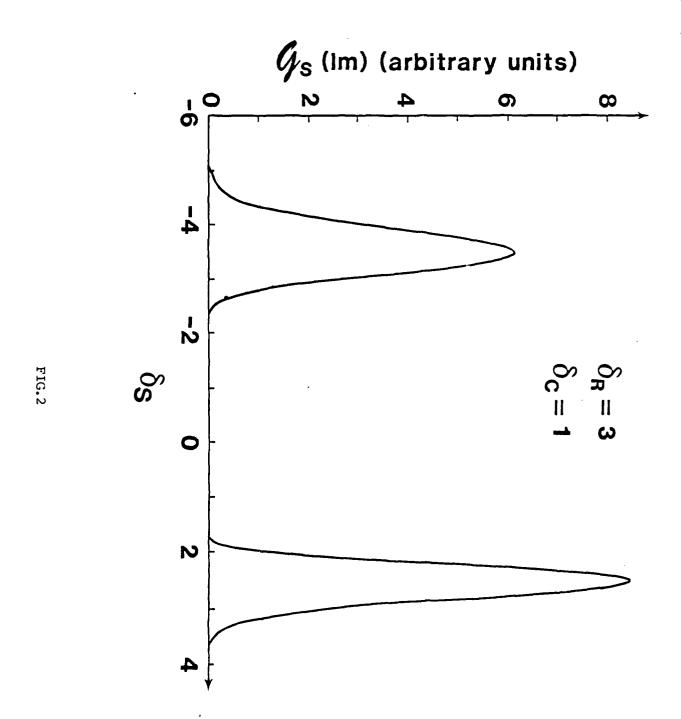


FIG.1



## APPENDIX B

## SOFT X-RAY AMPLIFICATION BY IONS SCATTERING FROM ANTIFERROMAGNETIC SURFACES

bу

A. Elçi

Institute for Modern Optics, Department of Physics and Astronomy, University of New Mexico, Albuquerque, New Mexico 87131, U.S.A.,

and

M.O. Scully

Max-Planck-Institut für Quantenoptik, Garching D-8046, BRD.

## abstract

We show that hydrogenic ions scattering from antiferromagnetic surfaces execute rapid Rabi oscillations in their rest frames, which can be used to amplify X-rays.

#### I. INTRODUCTION

The subject of the present paper was inspired by the free electron lasers. In free electron lasers, the laser action, or amplification, occurs at the wavelength  $\lambda$  = 2  $\lambda_{\alpha}/\gamma$  , where  $\lambda_{\alpha}$  is the wiggler wavelength of the magnetic selonoid and Y is the ratio of the electron's energy to its rest mass energy [1]. In principle, one can decrease  $\lambda$  either by increasing the electronic energy, or by decreasing the wiggler wavelength  $\lambda_{\alpha}$ . The latter is not very practical with the mechanical systems that are used in free electron lasers. There are, however, systems which are periodic on atomic scale, and which might be used as short wavelength wigglers for electrons or ions. A particularly interesting idea is to use antiferromagnetic crystals as wigglers for ions. These crystals have strong internal magnetic fields that are periodic with some lattice periodicity [2-4] and can be used to excite ions coherently. Excited ions can then decay radiatively and thus convert the virtual photons of the material magnetic field to real photons, just as in the case of free electron lasers.

In the following we discuss the physics of this problem in detail for hydrogenic ions. Specifically, we consider a beam of Be  $^{+3}$  ions scattering from the surface of an antiferromagnetic material like FeF or MnO. Be $^{+3}$  ions penetrate just below the surface of the material, travel for some distance L, then come out. They simultaneously interact with a soft X-Ray signal of frequency  $\hbar\omega_s \sim 30$  eV which matches the frequency  $\omega_{32}$  of the n=3 + n=2 transitions of Be $^{+3}$ . In Section II, we first consider the magnetization fields inside an

antiferromagnet and deduce the general form of the fields from a simple argument. We then obtain a simple set of equations of motion for the ionic amplitudes from the Schrodinger equation in the rest frame of an ion. These equations show that ions in their rest frame execute Rabi oscillations [5] between the states n=1 and n=3, if the ion velocity is properly adjusted. The Rabi frequency  $\Omega_{\rm p}$  is proportional to  $B_n$ , the amplitude of the magnetization of one of the sublattices of the antiferromagnet, and to v, the ion velocity.  $\Omega_n$  is on the order of  $4x10^{13}$  sec<sup>-1</sup> for  $v=10^9$  cm/sec, which is nearly four orders of magnitude larger than the spontaneous radiative decay rate of the n=3 states of  $Be^{+3}[6]$ . Since ions are actually moving with velocity v, the Rabi oscillations in the rest frame become spatial oscillations in the lab frame [7]. If L designates the spatial interval for half a Rabi cycle (i.e.,  $L_{R}$  is minimum distance required for an ion to travel in the material in order to get the ion fully excited into the n=3 states, if it is initially is in the ground state), then  $L_{\rm p}$  is on the order of  $10^{-4}$  cm. We also give a detailed discussion of the validity of the model. In Section III we calculate the gain of the signal field in the weak signal regime. Due to the coherent excitation of ions, the expression for the gain differs from the usual small-signal swept-gain [8], and has a number of novel features. It turns out that only for a very narrow region of detuning between  $\omega_{\varsigma}$  and  $\omega_{\,3\,2}$  can one get finite gain. The maximum gain is about 10 cm $^{-1}$  if the ionic density is on the order of 10½ cm $^{-3}$ . Section IV gives a further discussion of the results and our conclusions.

#### II. ANTIFERROMAGNETIC SURFACE SCATTERING

The schematic of ion scattering from an antiferromagnetic material is shown on Fig. 1. A beam of Be $^{+3}$  ions grazes the surface of an antiferromagnetic material at an angle  $\psi_1$  typically less than  $0.5^{\circ}$  [9], penetrates the surface, and, after traversing the material for some distance L, comes out. Simultaneously, the beam interacts with a soft X-ray signal of frequency  $\hbar\omega_{_{\rm S}}\sim 30$  eV, which corresponds to the transition frequency between the n=3 and n=2 levels of Be $^{+3}$ . This is the signal that we want to amplify. The signal propagates in the same direction as the ion beam. In this section we analyze this problem with the aid of a simple model. We first discuss the description of the fields of the antiferromagnetic material, then write a simple set of equations of motion for Be $^{+3}$  ion and explore the implications of these equations.

An antiferromagnetic crystal is composed of two interpenetrating identical sublattices which are shifted relative to each other by some spacing  $\vec{d}$ . In each sublattice, spins are localized at the lattice points and are parallel to each other; however, they are antiparallel to the spins of the other sublattice. The vector potential induced by the intrinsic magnetic moments of these spins has, therefore, the periodicity of the sublattice and can be written as

$$\vec{A}'_{M}(\vec{x}') = \sum_{\vec{G}} e^{i\vec{G}\cdot\vec{x}'} \vec{A}(\vec{G}) , \qquad (2.1)$$

where  $\vec{G}$  refers to the reciprocal lattice vectors of the sublattice. The primes are meant to indicate that these quantities are in the rest frame of the material medium. The presence of a surface breaks the translational symmetry normal to the surface and modifies (2.1). However, this modification is relatively simple once one has an expression for  $\vec{A}(\vec{G})$ . To see what the form of  $\vec{A}(\vec{G})$  is, we use the simplified picture of rigidly fixed spins. Each localized spin then gives rise to a magnetic dipole field, the superposition of which yields the overall vector potential:

$$\vec{A}_{M}'(\vec{x}') = g_{S} \mu_{B} \sum_{\vec{R}} \left[ \frac{\hat{u} \times (\vec{x}' - \vec{R})}{|\vec{x}' - \vec{R}|^{3}} - \frac{\hat{u} \times (\vec{x}' - \vec{R} - \vec{d})}{|\vec{x}' - \vec{R} - \vec{d}|^{3}} \right]$$
 (2.2)

where  $\textbf{g}_{\text{S}}$  is the gyromagnetic ratio of a given spin,  $\mu_{\text{B}}$  is the Bohr magneton, and

$$\vec{R} = \ell \vec{a} + m\vec{b} + n\vec{c}$$
,  $\ell, m, n = integers$ , (2.3)

are the lattice vectors of one of the sublattices.  $\mu$  designates the direction of magnetization of this sublattice. The Fourier transform of  $\overrightarrow{A}'(\overrightarrow{x}')$  can be written as

$$\vec{A}'_{M}(\vec{q}) = \int d\vec{r} e^{-i\vec{q} \cdot \vec{x}'} \vec{A}'_{M}(\vec{x}')$$

$$= (V/N) \left( \sum_{\vec{R}} e^{-i\vec{q} \cdot \vec{R}} \right) \vec{A}(\vec{q}) , \qquad (2.4)$$

where V is the volume, N is the number of the sublattice points, and  $\overset{\rightarrow}{\rightarrow}$  A(q) is defined by

$$\vec{A}(\vec{q}) = 2\pi N (1 - e^{-i\vec{q} \cdot \vec{d}}) \quad (V|\vec{q}|^2)^{-1} \{\hat{\varepsilon}_{+} [(\hat{\varepsilon}_{-} \cdot \hat{\mu})(\hat{z} \cdot \vec{q}) - (\hat{z} \cdot \hat{\mu})(\hat{\varepsilon}_{-} \cdot \vec{q})]$$

$$-\hat{\varepsilon}_{-} [(\hat{\varepsilon}_{+} \cdot \hat{\mu})(\hat{z} \cdot \vec{q}) - (\hat{z} \cdot \hat{\mu})(\hat{\varepsilon}_{+} \cdot \vec{q})]$$

$$+\hat{z} [(\hat{\varepsilon}_{+} \cdot \hat{\mu})(\hat{\varepsilon}_{-} \cdot \vec{q}) - (\hat{\varepsilon}_{-} \cdot \hat{\mu})(\hat{\varepsilon}_{+} \cdot \vec{q})] \} , \qquad (2.5)$$

and

$$\hat{\varepsilon}_{\pm} = \hat{x}' \pm i \hat{y}' . \tag{2.6}$$

When the number of sublattice points N is sufficiently large  $(N+\infty)$ , the sum over  $\overset{\rightarrow}{R}$  gives

$$\Sigma_{\vec{R}} = \frac{-i\vec{q} \cdot \vec{R}}{\vec{G}} = N\Sigma_{\vec{G}} \delta_{\vec{q},\vec{G}}^{\dagger}.$$
(2.7)

Inverting the Fourier transform, we then find

$$\vec{A}_{M}'(\vec{x}') = V^{-1} \Sigma_{\vec{q}} e^{i\vec{q} \cdot \vec{x}'} \vec{A}_{M}(\vec{q})$$

$$= \Sigma_{\vec{G}} e^{i\vec{G} \cdot \vec{x}'} \vec{A}(\vec{G}) . \qquad (2.8)$$

Consider now the special case in which  $\hat{\mu}$  is along  $\hat{x}$ , the reciprocal lattice is cubic,  $\hat{z}$  coincides with one of the lattice directions, and  $\hat{d}$  is midway along  $R_3$  such that

$$G_3 = (\frac{2\pi}{2d}) \times (integer) = \frac{\pi n}{d}.$$
 (2.9)

The factor

$$1 - e^{-i\vec{G} \cdot \vec{d}} = 1 - (-1)^n$$
 (2.10)

is nonzero only for odd n, and (2.8) becomes

$$\vec{A}_{M}'(\vec{x}') = 16\pi g_{S}^{\mu} B^{NV}^{-1} \sum_{\vec{G}} e^{i\vec{G}_{\perp} \cdot \vec{x}_{\perp}'} \times \sum_{n=0}^{\infty} [-\hat{y}' \frac{K_{n}}{(K_{n}^{2} + \vec{G}^{2})} \sin K_{n} z' - i\hat{z}' \frac{G_{2}^{\cos K_{n}} z'}{(K_{n}^{2} + \vec{G}^{2})}], \qquad (2.11)$$

where

$$K_n = \frac{\pi}{d}(2n+1)$$
 , (2.12)

$$\vec{G}_{\perp} = (\frac{2\pi m}{a}) \hat{x}' + (\frac{2\pi \ell}{b}) \hat{y}' , \qquad m, \ell = \text{integers.}$$
 (2.13)

Let the ion beam propagate in the  $\hat{z}$ -direction with a uniform cross section. A given ion then samples a vector potential that is approximately the average of (2.11) over the transverse coordinates. When such a spatial average is taken, only the  $\vec{G}_{\perp} = 0$  terms survive in (2.11), and the vector potential becomes

$$\vec{A}_{M}'(\vec{x}') = -\hat{y}'B_{0} \sum_{n=0}^{\infty} \frac{\sin K_{n}z'}{K_{n}},$$
 (2.14)

where B  $_0$  = 16 mNg  $_{\rm S}$   $_{\rm B}$  /V. The corresponding magnetic field is given by

$$\vec{B}_{M}' = \hat{x}' B_{0} \Sigma_{n=0}^{\infty} \cos K_{n} z' . \qquad (2.15)$$

When a surface is introduced, the basic modification occurs in (2.7). Let the surface be the y-z plane. We separate the sum in (2.7) into two parts, one part is a sum over the sublattice points on the y-z plane and the other part is a sum over the sublattice points along the  $\hat{x}$ -direction:

I

$$\Sigma_{\vec{R}} e^{-i\vec{q} \cdot \vec{R}} = (\Sigma_{m,n} e^{-i (mbq_2 + 2ndq_3)}) (\Sigma_{\ell} e^{-iq_1 a \ell})$$

$$= (N_S \Sigma_{G_2 G_3} \delta_{q_2, G_2} \delta_{q_3, G_3}) \rho_1 (q_1), \qquad (2.16)$$

where N<sub>S</sub> is the total number of sublattice points on the y-z plane and  $\rho_1(q_1)$  is the spin line-density fluctuation normal to the surface. Note that N<sub>S</sub> $\rho_1(0)$  is the total number of sublattice points. Eq. (2.8) is now replaced by

$$\vec{A}_{M}'(\vec{x}') = \Sigma_{q_{1}G_{2}G_{3}} e^{i(q_{1}x'+G_{2}y'+G_{3}z')\rho_{1}(q_{1})} \vec{A}(q_{1},G_{2},G_{3}).$$
 (2.17)

If one ignores the spatial variation in transverse coordinates and takes an average over them, (2.17) reduces to (2.14). If the beam is of small cross-section or of small thickness in the  $\hat{x}$ -direction, then

transverse variations are important and (2.8) and (2.17) give different results. In the following we ignore the transverse variations in the vector potential and use (2.14). This simplifies the algebra and allows us to concentrate on the essential physics of coherent excitation of ions.

In order to couple the electron of Be $^{+3}$  to the antiferromagnetic vector potential, one needs to transform the field given by (2.14) to the rest frame of an ion moving with velocity v in the  $\hat{z}$ -direction. In the rest frame of the ion,

$$\vec{A}_{M}(\vec{x}) = -\hat{y}\gamma B_{0} \quad \sum_{n=0}^{\infty} k_{n}^{-1} \sin(k_{n}z + \omega_{n}t)$$
, (2.18)

where

$$\gamma = (1 - v^2/c^2)^{-1/2}$$

$$k_n = \gamma K_n,$$

$$\omega_n = \gamma v K_n.$$

Thus the ion experiences a time-dependent field in its rest frame, whose harmonic frequencies can be matched to one of the ionic transition frequencies by varying the beam velocity. In particular, if one matches one of  $\omega_n$  to  $\omega_{31}$ , the transition frequency between the ground state and the n=3 states, then the magnetic field of the

material can resonantly pump the ions to the n=3 states. The ion then executes Rabi oscillations in its rest frame. The frequency matching  $\omega_n = \omega_{31}$  requires that

$$v = \frac{\omega_{31}}{\sqrt{\left(\frac{\omega_{31}}{c}\right)^2 + K_n^2}} = \frac{\omega_{31}}{\sqrt{\left(\frac{\omega_{31}}{c}\right)^2 + \frac{\pi^2}{d^2} (2n+1)^2}}$$
(2.19)

For Be<sup>+3</sup>,  $\omega_{31} \simeq 3 \times 10^{17} \text{sec}^{-1}$ . For an antiferromagnet like MnO, d  $\simeq 4 \text{Å}$ . Substituting these values into (2.19), one finds  $v(n=0) \approx 3 \times 10^9$  cm/sec and  $v(n=1) \approx 10^9$  cm/sec, etc. Finally, if the matching conditions are met for the beam and ions are coherently excited to the n=3 states, then some of these excited ions can decay into the n=2 states by emitting  $\hbar \omega_{32} \simeq 30$  eV photons, which will amplify the signal. It is clear that the proposed scheme uses the same type of excitation process as in the Okorokov effect [7,10-14].

In order to simplify the equations of motion we assume that pumping is strong but that the signal is weak. We first consider the Rabi equations for the n=1 and n=3 states coupled by the material field alone. We then use the solutions of these equations to calculate the amplitudes of the n=2 states which are coupled to the n=3 states by the weak signal. Let the ion wave function in its rest frame be

$$\psi(\vec{x},t) = \sum_{n \neq m} c \quad (t)e^{-iE_n t} u_{n \neq m}(\vec{x}), \qquad (2.20)$$

where  $u_{n l m}(\vec{x})$  are the hydrogenic wavefunctions for Be<sup>+3</sup>. We assume that the frequency matching condition is satisfied only for the n<sup>th</sup> harmonic of the material field. Neglecting the lifetimes and the counter-rotating terms, one then has the Rabi equations for n=1 and n=3 states of Be<sup>+3</sup>:

$$\frac{d}{dt} c_{100} = \frac{i e \gamma B_0}{m c k_n} e^{-i (\omega_{31} - \omega_n) t} \sum_{\ell', m'} P_{\gamma}(100; 3\ell' m' | k_n^2) c_{3\ell' m'}$$
(2.21)

$$\frac{d}{dt} c_{3\ell m} = \frac{i e y B_{2}}{m c k_{n}} e^{i (\omega_{31} - \omega_{n}) t} P_{y}(3\ell m; 100) - k_{n}^{2} c_{100}, \qquad (2.22)$$

where

$$\vec{P}(n\ell m; n'\ell' m' | \vec{k}) = \int d\vec{x} e^{-i\vec{k}\cdot\vec{x}} u_{n\ell m}^* (-i\vec{\nabla}) u_{n'\ell' m'}. \qquad (2.23)$$

We have kept just the  $\vec{p}$ . $\vec{A}$  terms in the above equations. Performing the indicated integral in (2.23) for n=3 and n=1 states, one obtains the matrix elements

$$P_y(3im;100|k^2) = -\frac{z}{a_B}(\delta_{m,1} + \delta_{m,-1})[\delta_{\ell,1}F_1(\xi)]$$

$$+ i \delta_{2,2} F_{2}(\xi)$$
, (2.24)

where Z is the nuclear charge of the hydrogenic ion and a is the Bohr radius. The other quantities are defined as follows:

$$\xi = \frac{4Z}{\sqrt{16Z^2 + 9k^2a_B^2}},$$
 (2.25)

$$F_{1}(\xi) = \frac{\xi^{4}(3+4\xi^{2})}{96} + \frac{\xi^{3}(1-\xi^{2})}{\sqrt{2\pi}(1+\xi)^{5/2}} \sum_{\ell=0}^{\infty} \frac{\Gamma(\ell-\frac{3}{2})\Gamma(\ell+\frac{5}{2})(1-\xi)^{\ell}}{\Gamma(\ell+\frac{7}{2})\Gamma(\ell+1)2^{\ell}},$$
(2.26)

$$F_{2}(\xi) = \frac{3\sqrt{5}}{40\sqrt{2}} \xi^{5} (1-\xi^{2})^{\frac{1}{2}} [(6-\sqrt{3})\xi^{2} + (\sqrt{3}-1)]$$
 (2.27)

Assuming that the ions are initially all in the ground state (i.e.,  $c_{130} = 1$ ,  $c_{32m} = 0$  at t = 0), the solutions of (2.21) and (2.22) are

$$c_{100}(t) = \frac{1}{(\Omega^+ - \bar{\Omega}^-)} (\Omega^+ e^{-i\Omega^- t} - \bar{\Omega}^- e^{-i\Omega^+ t}),$$
 (2.28)

$$e_{32m}(t) = \frac{e_Y B_y}{mck_n(x^+ - x^-)} P_y(32m; 100 | -k_n \hat{z}) \left(e^{ix^+ t} - e^{ix^- t}\right),$$
 (2.29)

where

$$\Omega^{\pm} = \frac{\omega_{31} - \omega_{n}}{2} \pm \sqrt{\left(\frac{\omega_{31} - \omega_{n}}{2}\right)^{2} + \Omega_{R}^{2}}, \qquad (2.30)$$

 $\Omega_{\,\mathrm{R}}$  is the Rabi frequency given by

$$\Omega_{R} = \frac{e \gamma B_0 D_0}{m c k_n} = \frac{e B_0 D_0 \gamma v}{m c \omega_{31}} , \qquad (2.31)$$

$$D_0 = \frac{Z\sqrt{2}}{a_B} \left[ (F_1(\xi_n))^2 + (F_2(\xi_n))^2 \right]^{\frac{1}{2}} . \tag{2.32}$$

When  $\omega_{31}=\omega_n$  exactly, the populations of upper and lower states oscillate with the Rabi frequency  $\Omega_R$ ,  $\Omega_R$  depends linearly on the amplitude  $B_0$  of the magnetic induction of the material. For antiferromagnets  $\text{FeF}_2$  and MnO,  $B_0\simeq 2x \cdot 10^7\text{G}$  and  $5.4 \times 10^6\text{G}$ , respectively [15]. Taking  $B_0=5 \times 10^6\text{G}$  as typical, and assuming that the zeroth harmonic  $\omega_0$  is matched to  $\omega_{31}$ , we find  $D_0\simeq 9 \times 10^7$  cm<sup>-1</sup> and  $\Omega_R\simeq 8.9 \times 10^{13}$  sec<sup>-1</sup> for Z=4. Thus if the ion moves for a distance of  $L_R=\pi v/\Omega_R$  in the material with constant velocity  $v=3 \times 10^9$  cm/sec, then the population of ions, which is initially in the ground state, is completely excited to the n=3 states. Table I lists some of the relevant numbers for matching  $\omega_{31}$  to some of the harmonics.

It follows from (2.24) that only certain states of the n=3 shell are coupled to the ground state. These are the states with  $\ell$ =1 and  $\ell$ =2, and m= $\pm$ 1. The coupling of  $\ell$ =2 states obviously arises from higher multipole transitions than the dipole, since the change in the

angular momentum  $\Delta \ell = 2$ . However,  $F_1(\xi)$  which corresponds to  $\Delta \ell = 1$  in (2.24) also has contributions from the higher multipoles. To be precise, the electric dipole transitions correspond to the limit  $\xi \neq 1$  of  $P_y$ . In this limit the sum in (2-26), as well as  $F_2$ , vanish, and the first group of terms in (2-26) yields  $F_1(1) \approx 0.1$ . The importance of the contribution of the higher multipoles depends sensitively upon the size of  $k_n$  via the relation (2-25).

Let us now consider the amplitudes of the n=2 states. Let the signal field be described by

$$\vec{A}_{S} = \hat{y}A_{S} \cos(k_{z}z - \omega_{S}t) . \qquad (2.33)$$

If the lifetime of the n=2 states is  $\Gamma_2$ , then

$$\left(\frac{d}{dt} + \Gamma_2\right) c_{2lm} = \frac{ieA_s}{mc} e^{-i(\omega_{32} - \omega_s)t}$$
 (2.34)

Substituting the solution for  $c_{3\ell,m}$  from (2.29) and integrating the result (assuming  $c_{2\ell,m}=0$  at t=0), one finds

$$c_{2\ell m}(t) = \frac{e^2 \gamma B_0 A_s}{m^2 c^2 (\Omega^+ - \Omega^-) k_n} \left[ \sum_{\ell' m'} P_{\nu}(2\ell m; 3\ell' m' | -k_s \hat{z}) P_{\nu}(3\ell' m'; 100 | -k_n \hat{z}) \right]$$

$$\times \left[ \frac{i \left(\Omega^{+} - \omega_{32} + \omega_{s}\right) t}{e^{-} + \omega_{32} + \omega_{s} - i \frac{\Gamma_{2}}{2}} - \frac{i \left(\Omega^{-} - \omega_{32} + \omega_{s}\right) t}{e^{-} + \omega_{32} + \omega_{s} - i \frac{\Gamma_{2}}{2}} \right]$$

(2.35)

We note that  $k_S$  is at least two orders of magnitude less than  $k_n$ , and therefore, one can let  $k_S \alpha_B \to 0$  and use the dipole approximation in the evaluation of the momentum matrix element with  $k_S$ . In this limit,

$$P_y(3lm;2l'm'|0) = -\frac{Z}{a_B} \delta_{m',0}(\delta_{m,1} + \delta_{m,-1})$$

$$\times \left[\delta_{\ell,1}\delta_{\ell',0}\sqrt{\eta_1} + \delta_{\ell,2}\delta_{\ell',1}\sqrt{\eta_2}\right]$$
 (2.36)

where  $n_1$  and  $n_2$  are pure numbers given by

$$\eta_1 = \frac{2^{13} \cdot 3^2}{5^{10}} \approx 7.55 \times 10^{-3},$$

$$\eta_2 = \frac{2^{18} \cdot 3^3}{5^{10}} \approx 0.72. \tag{2.37}$$

It is worth pointing out that the solution (2.35) indicates a Raman type coupling, since the amplitude is proportional to  ${\bf A}_{M-S}$  as in the Raman scattering.

In the preceding discussion we assumed that the velocity of the ion remains the same as at the time of the entry of the ion into the material. This is of course a gross approximation. As soon as the ion enters the surface region, it begins to lose energy, particularly to the electrons of the target material, and slows down. We can estimate the error involved in the assumption of constant ion velocity from the stopping power of electrons in solids, which depends on the speed of the ion, its charge number Z, and the charge number of the target atoms  $Z_T$  [16-18]. The stopping power scales as

$$S(Z,Z_T,v) = Z^2 S_p(Z_T,v) \left[\frac{Z^*(v)}{ZZ_p(v)}\right]^2$$
 (2.38)

where  $S_p$  is the stopping power when the projectile is a proton, and  $Z^*(v)$  and  $Z_p(v)$  are the effective charges of the ion and the proton, respectively, in the material [16]. These effective charges arise from the screening effects due to the electrons of the material and

are dependent on the projectile speed [20-22]. Let the target atoms be Mn and v =  $10^9$  cm/sec. For this speed  $S_p \approx 10^{-14} \text{eV.cm}^2/\text{atom}$ ,  $Z^*/(ZZ_n) \approx 0.6$ , and therefore,  $S \approx 6x10^{-14} \text{ eV.cm}^2/\text{atom.}$  The number of target atoms per unit volume in MnO is on the order of N  $_{_{\rm T}}$   $\approx$  10  $^2$   $^2$  cm  $^{-3}$  , which translates into an energy loss per unit length N  $_{\!\scriptscriptstyle T} \text{S}~\approx~9\text{x}\text{10}^{~8}$ eV/cm. If the distance travelled by the ion is  $L_{_{D}}$   $\approx$  0.8  $\mu\text{m},$  which is the minimum necessary distance needed to get the ion completely excited into the n=3 states, then the total energy loss is  $\delta E$  =  $N_{\rm T} S L_{\rm p}^{\,\,\approx}$  50 keV. The relative decrease in speed is given by  $\delta v/v = \delta E/2E \sim 4x10^{-2}$ . These numbers show that the constant velocity assumption is not too bad, in the sense that the error in velocity is on the order of 4% per Rabi cycle. We should point out these are bulk estimates, and are therefore likely to be much larger than the corresponding surface rates. As the ion slows down,  $\omega_{3\,1}$  and  $\omega_{n}$  become detuned. In terms of the stopping power, the detuning from the pump frequency is given by

$$\Delta_{n} = k_{n} \delta v = \frac{v N_{T} L_{R} k_{n} S}{2E}$$
 (2.39)

Estimates of  $\hbar \Delta_0$  and  $\hbar \Delta_1$  are given in Table I.

Another important assumption in the above discussion is that  $Be^{+3}$  ion remains  $Be^{+3}$  inside the material medium, that is, it keeps to its original charge state. There is some conflicting opinion and evidence on the states of an ion or atom while it travels inside a solid

material [23-27]. The projectile may lose its original electron and be completely stripped, or it may capture electrons, losing them only while leaving the solid material [21]. At any rate, the charge states of a projectile may not be entirely determined. In view of this situation, how reliable is the above assumption and the consequent analysis? There are three possible answers to this question. First, the charge state of the ion is clearly dependent on the ion speed. At low speeds the charge state appears to remain unchanged with relatively high probability [24]. One can, therefore, reduce the ionic speed by matching the relevant ion transition frequency to a higher harmonic of the material field. There is probably some trade off point in this procedure, since the Rabi frequencies of the higher harmonics of the material fields are lower, leading to larger energy losses. Second, even for swift ions, the ion spends some of its time inside the solid in its original charge state due to repeated capture and loss of electrons of the medium. From the cross sections for electron capture or loss [14,28,29], one can estimate the probability of the ion being in its original charge state, and accordingly find the expected values of the desired quantities from this probability. This picture of ions being partially in their original charge states is supported by the correlations observed between ions and a coterie of electrons that are emitted secondarily [30]. Finally and third, in the treatments of the Okorokov effect, one uses the picture of the projectile being in its entry charge state [7,10,11]. One then

calculates the probability of excitation of the projectile as a function of its speed. The model apparently works, and there is a fair agreement with experiment [12-14].

The cross sections for the electron capture and loss are on the order of  $\sigma_{\rm c} \simeq \sigma_{\ell} \simeq 10^{-19}~{\rm cm}^2$  [14,24,28]. The target mobile electron density for MnO is on the order of N<sub>e</sub>  $\simeq 10^{17}~{\rm cm}^{-3}$ . It follows that the probability of the ion changing its charge state is roughly  $(\sigma_{\rm c} + \sigma_{\ell}) N_{\rm e} L_{\rm R} \simeq 10^{-6}$  per half Rabi cycle.

## III. GAIN FOR THE X-RAY SIGNAL

In order to calculate the gain for the signal field, we follow the well known procedure used in traveling wave amplifiers and free electron lasers [31]. We calculate the transverse charge current density of the ionic electrons which oscillates as e  $^{\rm s}$ , and substitute it into the wave equation. The wave equation yields a dispersion relation which requires either temporal or spatial growth. The latter yields the small signal gain for the X-ray signal.

The  $n=3 \rightarrow n=2$  transitions cause a particle current density that is given by

$$J_{y}(k_{s}^{2},t) = \frac{\hbar e^{-i\omega_{32}t}}{m} \sum_{\ell m \ell' m'} c_{2\ell m}^{*}(t) P_{y}(2\ell m; 3\ell' m' \mid 0) c_{3\ell' m'}(t) . \qquad (3.1)$$

It follows from (2-29) and (2-35) that the component which oscillates  $i\omega_s^t$  as e  $i\omega_s^t$ 

$$j_{y}^{(s)} = \frac{e^{3} \hbar \gamma^{2} B_{o}^{2} A_{s}^{D} I_{1}^{4}}{m^{4} c^{3} (\Omega^{+} - \Omega^{-})^{2} k_{n}^{2}} \left[ \frac{1}{\Omega^{+} - \omega_{32}^{+} \omega_{s}^{+} \frac{i \Gamma_{2}}{2}} + \frac{1}{\Omega^{-} - \omega_{32}^{+} \omega_{s}^{+} \frac{i \Gamma_{2}}{2}} \right]$$
(3.2)

where

$$D_{1}^{4} = \sum_{\ell m \ell' m' \ell' m''} P_{y}(2\ell m; 3\ell' m' \mid 0) P_{y}(3\ell'' m''; 2\ell m \mid 0)$$

$$\times P_{y}(3l'm';100|-k_{n}\hat{z})P_{y}(100;3l''m''|k_{n}\hat{z}). \tag{3.3}$$

Using (2.24) and (2.36),  $D_1$  becomes

$$D_{1} = \frac{Z}{a_{p}} \left[ \eta_{1}(F_{1}(\xi_{n}))^{2} + \eta_{2}(F_{2}(\xi_{n}))^{2} \right]^{\frac{1}{4}}$$
(3.4)

 $j_s^{(s)}$  is the transverse current density of just one ion. If there are  $N_i$  ions per unit volume in the ion beam moving with velocity v, then the total transverse charge density component is given by  $ej_s^{(s)}N_i$ , and the wave equation becomes

$$\left(\frac{\partial^2}{\partial z^2} - \frac{1}{c^2} \frac{\partial^2}{\partial t^2}\right) \vec{A}_s^+ = -\hat{y} \left(\frac{4\pi e N_i}{c}\right) j_y^{(s)} e^{ik_s^2 - i\omega_s t}$$
(3.5)

where  $\vec{A}_s = \hat{y} \hat{A}_s e^{-i\omega_s t}$  is the positive frequency component of the signal. Performing the indicated operations in (3.5), one obtains the dispersion relation

$$-k_{s}^{2} + \frac{\omega_{s}^{2}}{c^{2}} = -\frac{4\pi e^{4} \aleph_{1}^{2} \aleph_{0}^{2} \aleph_{1}^{1} \aleph_{1}^{4}}{m^{4} c^{4} k_{n}^{4} (\Omega^{+} - \Omega^{-})^{2}} \left[ \frac{\Omega^{+} - \omega_{32} + \omega_{s} - i \frac{\Gamma_{2}}{2}}{(\Omega^{+} - \omega_{32} + \omega_{s})^{2} + \frac{1}{4} \Gamma_{2}^{2}} + \frac{\Omega^{-} - \omega_{32} + \omega_{s} - i \frac{\Gamma_{2}}{2}}{(\Omega^{-} - \omega_{32} + \omega_{s})^{2} + \frac{1}{4} \Gamma_{2}^{2}} \right]$$
(3.6)

For an amplifier, one lets the frequency be real but allows the wavevector to have an imaginary part in order to determine the growth or the decay of the signal in space. Thus, let

$$k_s = \frac{\omega_s}{c} - ig . ag{3.7}$$

Here a positive g means that the signal wave grows as  $\exp(gz)$  and thus g is the gain due to the ions moving with velocity v. g is generally quite small compared to  $\frac{1}{2}$  /c; therefore,

$$k_s^2 = \frac{\omega_s^2}{c^2} - 2ig \frac{\omega_s}{c} - g^2 \simeq \frac{\omega_s^2}{c^2} - 2ig \frac{\omega_s}{c}$$
, (3.8)

and

$$g = \frac{\pi N e^{4} \gamma^{2} B_{0}^{2} N_{i} D_{1}^{4}}{\omega_{s}^{m} k_{n}^{2} c^{3} (\Omega^{+} - \Omega^{-})^{2}} \left[ \frac{\Gamma_{2}}{(\Omega^{+} - \omega_{32}^{+} \omega_{s})^{2} + \frac{1}{4} \Gamma_{2}^{2}} + \frac{\Gamma_{2}}{(\Omega^{-} - \omega_{32}^{+} \omega_{s})^{2} + \frac{1}{4} \Gamma_{2}^{2}} \right]$$
(3.9)

In the limit  $\Gamma_2 \rightarrow 0$ , the terms bracketed are reduced to a pair of delta functions:

$$g = \frac{2\pi^{2} \text{Ne}^{4} \gamma^{4} B_{0}^{2} N_{1} D_{1}^{4}}{\omega_{s}^{m} k_{n}^{2} c^{3} (\Omega^{+} - \Omega^{-})^{2}} \left[ \delta(\Omega^{+} - \omega_{32} + \omega_{s}) + \delta(\Omega^{-} - \omega_{32} + \omega_{s}) \right] .$$
(3.10)

In an ionic beam, ions will in general have a dispersion of velocities around an average velocity  $\mathbf{v}_0\hat{\mathbf{z}}$ . Ions moving with slightly different velocities will be differently tuned to the material field and contribute to the overall gain with different strengths. Assuming that the beam of Be<sup>+3</sup> has a narrow Gaussian distribution of velocities in the  $\hat{\mathbf{z}}$ -direction described by

$$\rho_{i}(v) = \frac{1}{\Delta v v^{2}} \exp\left[-\frac{(v-v_{0})^{2}}{\Delta v^{2}}\right],$$
(3.11)

the net gain is given by

$$\bar{g} = p_{CS} \int_{-\infty}^{+\infty} dv g(v) \rho_i(v)$$
,
(3.12)

where we have multiplied the average over the velocity profile with  $\mathbf{p}_{\text{CS}}$  = the probability of the Be<sup>43</sup>ion being in the charge state +3 (roughly 0.5). If  $\Gamma_2 < \mathbf{k}_n \Delta \mathbf{v}$ , one can use the simpler form of the gain given by (3.10). The integral in (3.12) is then readily done and one finds

$$\bar{g} = \frac{4\pi^{2} \pi^{4} e^{4} \gamma^{2} B_{0}^{2} N_{i} D_{1}^{4} P_{CS} \rho_{i} (v_{t})}{\omega_{s} k_{p}^{3} m^{4} c^{3} [(\omega_{s} - \omega_{32})^{2} + \Omega_{R}^{2}]},$$
(3.13)

where  $\mathbf{v}_{t}$  is some tuning velocity given by

$$v_t = \frac{\omega_s - \omega_{32}}{\Omega_R} - \frac{\Omega_R^2}{k_n(\omega_s - \omega_{32})} + \frac{\omega_{31}}{k_n}$$
 (3.14)

The expression (3.13) for  $\overline{g}$  can be put into a more recognizable form in order to compare it with the usual small signal swept-gain [8]. Define an "effective" Einstein A-coefficient

$$A_{eff} = \frac{2\omega_{s}^{2}u_{eff}^{2}}{3\pi c^{3}}$$
,  $\mu_{eff} = \frac{\sqrt{2} e MD_{1}^{2}p_{CS}}{m\omega_{32}D_{0}}$ , (3.15)

and the Doppler broadening half-width

$$\Delta\omega_{D} = (k_{n}\Delta v) 2\sqrt{\ell n} 2 \qquad . \tag{3.16}$$

(3.13) can be written

$$\overline{g} = \frac{3}{2} \left( \frac{\ln 2}{\pi} \right)^{1/2} \lambda_{s}^{2} \left( \frac{A_{eff}}{\Delta \omega_{D}} \right) N_{i} \left[ \frac{\Omega_{R}^{2}}{(\omega_{s} - \omega_{32})^{2} + \Omega_{R}^{2}} \right] \exp \left[ -\frac{(k_{n} v_{t} - \omega_{n}^{0})^{2} 4 \ln 2}{(\Delta \omega_{D})^{2}} \right],$$
(3.17)

where  $\omega_n^{\ 0}=k_n^{\ v_0}$ . It is seen that (3.17) differs from the usual gain expression in two ways. First, instead of the usual population inversion factor  $\Delta N=N_{\rm excited}^{\ -N_{\rm ground}^{\ }}$ , (3.17) has a density factor that refers to the total density of the ions modulated by the Rabi frequency and the detuning of  $\omega_s$  from  $\omega_{32}$ . In other words, the entire collection of ions participate in the excitation and emission processes. This is the origin of the phrase "coherent excitation." Second, (3.17) has a Doppler factor that has complicated  $\omega_s$ -dependence and goes to zero exactly at  $\omega_s=\omega_{32}$ . Thus the following correspondences exist when compared with the usual gain:

$$\exp \left[-\frac{(k_{\rm n}v_{\rm t}^{-}\omega_{\rm n}^{0})^{2}4\ell_{\rm n2}}{(\Delta\omega_{\rm D})^{2}}\right] + \exp \left[-\frac{(\omega_{\rm s}^{-}\omega_{\rm 32})^{2}4\ell_{\rm n2}}{(\Delta\omega_{\rm D})^{2}}\right],$$

$$N_{i} \left[ \frac{\Omega_{R}^{2}}{(\omega_{s}^{-\omega_{32}})^{2} + \Omega_{R}^{2}} \right] \rightarrow (N_{\text{excited}}^{-N_{\text{ground}}})$$
 (3.18)

The fact that the excitation is coherent is quite important, since essentially all ions in the beam contribute to the gain and the question of population inversion does not arise. Instead, the question becomes one of tuning relative to the Rabi frequency, which determines the effective ion density that contributes to the gain. Since this tuning should be achieved with relative ease, coherent excitation has a significant advantage over the conventional incoherent excitation of a laser medium.

An interesting feature of (3.13) is that the gain scales as some frequency detunings per unit Rabi frequency, and the maximum possible gain is independent of the size of magnetization. To be precise, we can normalize the ion beam density to N  $_{io}$  = 10  $^{14}$  cm  $^{-3}$  and write  $\overline{g}$  as

$$\overline{g} = g_0(\frac{N_i}{N_{i0}}) (\frac{v_0}{\Delta v}) (\frac{1}{1+\delta^2}) \exp \left[-\frac{(1-\delta\delta_p - \delta^2)^2}{v^2 \delta^2}\right] , \qquad (3.19)$$

where

$$g_0 = \frac{4\pi^{3/2} e^2 M N_{i0} D_1^4 P_{CS}}{m^2 v_0 ck_n \omega_s D_0^2},$$
(3.20)

$$\delta = (\omega_s - \omega_{32}) / \Omega_R , \qquad (3.21)$$

$$\delta_{p} = (\omega_{31} - k_{n} v_{0}) / \Omega_{R}$$
 (3.22)

and

$$w = k_n \Delta v / \Omega_R \tag{3.23}$$

As seen from Table I,  $g_0 \sim 10^{-4}~cm^{-1}$  for  $lo_W$  harmonics. Due to slowing down of ions, one can expect  $\delta_p$  to be on the order of  $\Delta_n/\Omega_R$  which is a number generally larger than 10. Because of the factor  $(\mathbf{v}_0/\Delta_V)$ , it is desirable to have as small  $(\Delta v/\mathbf{v}_0)$  as possible. By pushing to the limits of the present ion beam technology it is possible to get a high velocity resolution on the order of  $(\Delta v/\mathbf{v}_0) \sim 10^{-5}$ . W is then 0.04 and 0.15 for the zeroth and first harmonic frequency matching, respectively. Clearly, the maximum gain is obtained when  $\delta$  is approximately given by

$$\delta_{\rm m} = -\frac{1}{2} \delta_{\rm p} + \sqrt{\frac{1}{4} \delta_{\rm p}^2 + 1} \simeq \frac{1}{\delta_{\rm p}}$$
 (3.24)

which makes the exponential factor unity. For  $\delta = \delta_m$ ,

$$\bar{g} = g_0 \left( \frac{N_i}{N_{i0}} \right) \left( \frac{v_0}{\Delta v} \right) \left( \frac{\delta_p + \sqrt{4 + \delta_p^2}}{2\sqrt{4 + \delta_p^2}} \right) 
\approx g_0 \left( \frac{N_i}{N_{i0}} \right) \left( \frac{v_0}{\Delta v} \right) \left( 1 - \frac{1}{\delta_p^2} \right) .$$
(3.25)

Thus, for  $N_i = 10^{14} \text{ cm}^{-3}$  and  $2\text{v/v}_0 = 10^{-5}$ ,  $\overline{g} \sim 10 \text{ cm}^{-1}$  which is a significant gain. Note that the gain is confined to a narrow tuning

$$\exp \, \sim \, 1 - \frac{\delta_{p}^{4}}{4} \, (\delta - \frac{1}{\delta_{p}})^{2} \, .$$
 (3.26)

It follows that for  $\delta = \delta_m \sim (w^2/\delta_p^2)$  which is on the order of  $10^{-4}$  or less,  $\overline{g}$  is substantially reduced.

## IV. CONCLUSION

With slight modifications the preceding analysis applies to non-hydrogenic ions and neutral atoms. We therefore conclude that ion-antiferromagnetic surface scattering is a promising and quite general scheme for X-ray amplification and X-ray lasers.

Note that for  $N_i = 10^{14}$  cm<sup>-3</sup>, the charge current density of the ions is  $|e|N_i v_0 Z \sim 6.5 \times 10^4$  amp/cm<sup>2</sup> for  $v=10^9$  cm/sec. Or in terms of the power contained in the ionic beam,  $P_i = \frac{1}{2} N_i M_{ion} v^3 \sim 9 \times 10^4$  MW/cm<sup>2</sup>. Clearly, in an actual experiment, the particle beam would have to be pulses of a nearly neutral plasma beam.

Admittedly the beam densities required to achieve  $\overline{g} \sim 10$  cm<sup>-1</sup> are high [32]. However, presently there are sources for Li ions that can yield such high densities [33]. Furthermore, ion beams can be focused on the surface to obtain high densities [34]. The experimental difficulties may lie with combining a high velocity resolution with high densities.

Another possibility is to increase the effective range of interaction L between the signal and the excited ions. The actual  ${\rm e\,f\,f}$ 

amplification of the signal amplitude is given by  $e^{SL_{eff}}$ . If  $L_{eff}$  is sufficiently large, much smaller  $\tilde{g}$  can be tolerated to obtain significant signal amplification.  $L_{eff}$  can be increased in a storage ring configuration, where both the particle beam pulses and the X-ray signal repeatedly scatter from surfaces that are placed in a circular fashion (with a large radius to allow for grazing angle scatterings). Between surface scatterings, the velocity spread of the particle beam can be adjusted by means of external fields.

Finally, the preceding analysis shows that the antiferromagnetic surface scattering is an extremely efficient pump mechanism to create an inverted population.  $\Omega_{\rm R}$  which determines the rate of production of the excited states is four to five orders of magnitude larger the radiative lifetimes of the relevant excited states.

Acknowledgement: We thank C. Rau and D. M. Wolfe for useful discussions on the problem. This work was supported by the Air Force Office of Scientific Research and Max-Planck-Gesellshaft zur Förderung der Wissenschaften e.V.

### References

- W. H. Louisell, J. F. Lam, D. A. Copeland, and W. B. Colson, Phys. Rev. A 19 (1979) 288.
- 2. T. Nagamiya, K. Yosida, and R. Kubo, Adv. in Phys. 4 (1955) 1.
- 3. A. Arrott, "Antiferromagnetism in Metals", in Magnetism, Vol.

  IIB, eds. G. T. Rado and H. Suhl (Academic Press, New York 1965).
- V. Jaccarino, "Nuclear Resonance in Antiferromagnets," in <u>Magnetism</u>, Vol. IIA, eds. G. T. Rado and H. Suhl (Academic Press, New York 1966).
- 5. M. Sargent, M. O. Scully and W. Lamb, Jr. Laser physics (Addison-Wesley, Reading, Massachusetts 1974).
- 6. M. Mizushima, Quantum Mechanics of Atomic Spectra and Atomic Structure (W. A. Benjamin, Inc., New York 1970), Sec 5 6.
- 7. J. Kundo, J. Phys. Soc. Jap. 36 (1974) 1406.
- 8. R. Bonifacio, F. Hopf, P. Meystre, and M. O. Scully, "Theory of a short wavelength swept-gain amplifier," Physics of Quantum Electronics, Vol. 3, eds. S. F. jacobs, M. O. Scully, M. Sargent, and C. D. Cantrell (Addision-Wesley, Reading, Massachusetts 1976).
- 9. For grazing angle scatterings the damage to the material is minimized. See C. Rau and R. Sizmann, "Electron spin polarization at ferromagnetic single crystalline Nickel surfaces determined through electron capture by scattered Deuterons," in Atomic Collisions in Solids, Vol. 1, eds. S. Datz, B. R. Appleton, and C. D. Moak (Plenum Press, New York 1975). In order

- to minimize energy losses,  $\psi_{\hat{\mathbf{i}}}$  should be chosen to satisfy channeling criteria; see Ref. 34.
- 10. V. V. Okorokov, JETP Letts. 2 (1965) 111.
- 11. S. Shindo and Y. H. Ohtsuki, Phys. Rev. B14 (1976) 3929.
- 12. V. V. Okorokov, D. L. Tolchenkov, I. S. Khizhnyakov, Yu. N. Cheblukov, Yu. Ya. Lapitsky, G. A. Iferov, and Yu. N. Zhukova, Phys. Letts. 43A (1973) 485.
- 13. M. J. Gaillard, J. C. Poizat, J. Remillieux, and M. L. Gaillard, Phys. Letts. 45A (1973) 306.
- 14. S. Datz, C. D. Moak, O. H. Crawford, H. F. Kruse, P. F. Dittner, J. Gomez del Campo, J. A. Biggerstaff, P. D. Miller, P. Hvelpund, and H. Knudsen, Phys. Rev. Letts. 40 (1978) 843.
- 15. These estimates are obtained from antiferromagnetic resonance frequencies. See. C. Kittel, Introduction to Solid State Physics (fourth edition, John Wiley and Sons, New York 1971), Ch. 17.
- 16. B. S. Yarlagadda, J. E. Robinson, and W. Brandt, Phys. Rev. <u>B17</u> (1978) 3473.
- 17. E. C. Montenegro, S. A. Cruz, and C. Vargas-Aburto, Phys. Letts.

  92A (1982) 195.
- 18. K. Dettmann, "Stopping power of fast channelled protons in the impact parameter treatment of atomic collisions," in <u>Atomic Collisions in Solids</u>, Vol. 1, eds. S. Datz, B. R. Appleton, and C. D. Moak (Plenum Press, New York 1975).
- K. A. Brueckner, L. Senbetu, and N. Metzler, Phys. Rev. <u>B</u> 25 (1982) 4377.

- 20. P. M. Echenique, R. H. Ritchie, and W. Brandt, Phys. Rev. <u>B</u> 20 (1979) 2567.
- 21. W. Brandt, "Ion screening in solids," in <u>Atomic Collisions in Solids</u>, Vol. 1, eds. S. Datz, B. R. Appleton, and C. D. Moak (Plenum Press, New York 1975).
- 22. W. Brandt, R. Laubert, M. Mourino, and A. Schwartchild, Phys. Rev. Lett. 30 (1973) 358.
- 23. H. D. Betz, Rev. Mod. Phys. 44 (1972) 465.
- 24. S. Datz, Nucl. Inst. Meth. 132 (1976) 7.
- 25. H. D. Betz, Nucl. Inst. Meth. 132 (1976) 19.
- 26. H. D. Betz and L. Grodzins, Phys. Rev. Lett. 25 (1970) 211.
- 27. C. D. Moak, S. Datz, B. R. Appleton, J. A. Biggerstaff, M. D. Brown, H. F. Krause, and T. S. Noggle, Phys. Rev. <u>B</u> 10 (1974) 2681.
- 28. S. J. Pfeifer and R. E. Olson, Phys. Letts. 92A (1982) 175.
- 29. W. Brandt and R. Sizman, Phys. Letts. 37A (1971) 115.
- 30. K. G. Harrison and M. W. Lucas, Phys. Letts. 33A (1970) 142.
- 31. N. M. Kroll, "The free electron laser as a travelling wave amplifier," in Physics of Quantum Electronic, Vol. 5, eds. S. F. Jacobs, M. Sargent, and M. O. Scully (Addison-Wesley, Reading, Massachusetts 1978).
- 32. For a review of intense pulsed ion beams, see S. Hamphries, Jr., Nucl. Fusion 20 (1980) 1549.

- 33. J. S. Helman, C. Rau and C. F. Bunge, "X-ray laser implementation by means of a strong source of high-spin metastable atoms," to be published in Phys. Rev. A (Jan. 1983).
- 34. For a discussion of surface reflection of ions, see R. Sizmann and C. Varelas, Nucl. Inst. and Meth. 132 (1976) 633.

Table I. Estimates of some parameters for Be<sup>+3</sup> ions and selected harmonics.

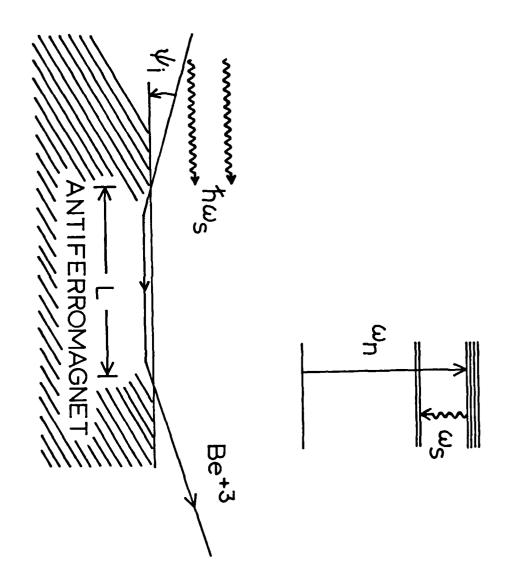
h∆(eV)	0.1	<b>∞</b>	*
$\frac{L_{\rm R}(10^{-4}{\rm cm})}{g_0(10^{-5}{\rm cm}^{-1})}$	5.9	14.8	15.8
$L_{\rm R}(10^{-4}{\rm cm})$	1.1	0.8	1.7
$\frac{D_1(10^8 \text{cm}^{-1})}{D_1(10^{13} \text{sec}^{-1})}$	8.9	4.1	9.0
$D_1 (10^8 \text{cm}^{-1})$	1.5	2.2	1.4
$D_0(10^8 \text{cm}^{-1})$	0.9	1.2	0.5
$k_n(10^8 cm^{-1})$	1.0	3.0	9.0
v(10 <sup>9</sup> cm/sec)	3.0	1.0	0.3
<b>=</b>	0	-	4

 $^{\star}$  For this speed  $S_{\rm p}$  is near its maximum, and (2.38) suggests that the ion will be

completely stopped in the material.

FIGURE CAPTION

Fig.1 The schematic of the antiferromagnetic surface scattering.



# Magnetic surface-induced spin-flip transitions in singlet O<sub>2</sub>

## A. Elçi and M. S. Zubairy

Institute for Modern Optics, Department of Physics and Astronomy, University of New Mexico, Albuquerque, New Mexico 87131

#### P. Avizonis and M. O. Scully

U.S. Air Force Weapons Laboratory, Kirtland Air Force Base, Albuquerque, New Mexico 87117 (Received 22 September 1983)

We show that a sizable electric dipole moment can be induced between the  ${}^{3}\Sigma_{z}^{-}$  and  ${}^{1}\Delta_{z}$  states of  $O_2$  when oxygen is placed in an environment of particles with spin  $\frac{1}{2}$ .

#### I. INTRODUCTION

In diatomic molecules formed by the group-VI elements (e.g., O<sub>2</sub>,S<sub>2</sub>,SO) and by a combination of group-V and group-VII elements (e.g., NF,NCl,PCl), the spin selection rule  $\Delta S = 0$  leads to highly metastable states which can be very useful for high-power laser systems. A particularly well-known case is the  ${}^{1}\Delta_{z}$  state of oxygen molecule. Owing to spin and parity symmetry, its decay to the ground state  ${}^{3}\Sigma_{g}^{-}$  is highly forbidden with spontaneous decay rate  $2.6 \times 10^{-4}$  sec<sup>-1,2</sup> This fact is used in oxygen-iodine lasers to store the energy in the  ${}^{1}\Delta_{g}$  state of  $O_{2}$ , which is then collisionally transferred to atomic iodine via the reaction  $O_2(^1\Delta_g) + I(^2P_{3/2}) \rightarrow O_2(^3\Sigma_g^-) + I(^2P_{1/2})$ . Radiative emission takes place on the iodine through its decay back to  ${}^{2}P_{1/2}$ , which is a magnetic-dipole-allowed transition with a spontaneous transition rate 7.7 sec<sup>-1.3</sup> The frequencies of  ${}^{1}\Delta_{g}$ - ${}^{3}\Sigma_{g}^{-}$  and  ${}^{2}P_{1/2}$ - ${}^{2}P_{3/2}$  transitions match, which makes it possible to resonantly transfer energy between O<sub>2</sub> and I. In practice it is difficult to find such matching partners for other metastable systems which might have applications to high-power lasers. For this reason, as well as to avoid losses involved in collisional energy transfers, there has long been an interest in obtaining radiative emissions directly from the excited metastable system itself. To achieve this it is necessary to break the various symmetries of the metastable system. For instance, the parity symmetry can be broken by applying static electric field or nonresonant radiation, or by means of nonresonant collisions with another molecular or atomic system.4-6 On the other hand, breaking of the spin symmetry requires application of either spatially varying magnetic field as in a Stern-Gerlach device, or a direct spin-spin coupling to another system.

In this paper we consider the latter possibility for the singlet oxygen, where the electrons of O2 are coupled to a set of external spins through their internal magnetic moments. This situation can be realized by putting  $O_2(^1\Delta_{\bullet})$ in contact with a magnetic surface as discussed in Sec. III. By means of a simple perturbation analysis, we show that both parity and spin symmetries are broken and that an electric dipole moment is induced between  ${}^{1}\Delta_{g}$  and  ${}^{3}\Sigma_{g}^{-}$ which is on the order of  $10^{-2}$  D. This implies a spontane-

ous transition rate of about  $10^2 \, \mathrm{sec}^{-1}$ , which is nearly 6 orders of magnitude larger than the corresponding rate for isolated O2.

## II. INDUCED DIPOLE MOMENTS

Consider the situation illustrated in Fig. 1. To simplify the problem, we take into account just two of the valence electrons of  $O_2$ . These electrons are coupled to spin- $\frac{1}{2}$ particles at sites  $R_{\lambda}$  ( $\lambda = 1, 2, ..., N$ ) by means of magnetic dipole-dipole interactions

$$V = 2g\mu_B^2 \sum_{\lambda=1}^N \sum_{j=1,2} \frac{1}{|\vec{R}_{\lambda} - \vec{x}_j|^5} [3\vec{\sigma}_j \cdot (\vec{R}_{\lambda} - \vec{x}_j) \times \vec{\sigma}_{\lambda} \cdot (\vec{R}_{\lambda} - \vec{x}_j) \times |\vec{R} - \vec{x}_j|^2 \vec{\sigma}_j \cdot \vec{\sigma}_{\lambda}],$$
(1)

where  $\mu_B$  is the Bohr magneton,  $\vec{\sigma}_{\lambda}$  are the Pauli spin operators of the external spins, and g is their gyromagnetic ratio.  $\vec{\sigma}_1$  and  $\vec{\sigma}_2$  are the Pauli spin operators for the O<sub>2</sub> electrons. In the following, we assume that the external spins and O2 are fixed in space and ignore all other interactions between the two systems.

To first order, the perturbed states of the combined system is given by

$$|a,M\rangle_{V} = |a,M\rangle + \sum_{\substack{a' \neq a, \\ M' \neq M}} |a',M'\rangle \frac{\langle a',M' | V | a,M\rangle}{E_{a} + W_{M} - E_{3'} - W_{M'}},$$

(2)

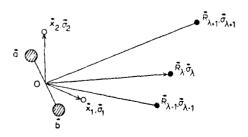


FIG. 1. Diagram for the notation of the coordinates.

<u>29</u>

where  $|a,M\rangle = |a\rangle \otimes |M\rangle$ ,  $|a\rangle$  and  $E_a$  designate the unperturbed states and energies of  $O_2$ , and  $|M\rangle$  and  $W_{M'}$  the unperturbed states and energies of the external spin system. Each M refers to a set of N parameters,  $|M\rangle = |s_1, s_2, \ldots, s_N\rangle$  where  $s_\lambda$  can take on only two values  $\pm 1$ , corresponding to spin up and down at the  $\lambda$ th site. It follows from (2) that the induced matrix elements of the dipole moment operator  $\vec{D}^{op} = -e(\vec{x}_1 + \vec{x}_2)$  are

$$V(a,M \mid \vec{\mathbf{D}}^{\text{op}} \mid a',M')_{V} = \delta_{MM'} \sum_{a''} \left[ \frac{\langle a,M \mid \vec{\mathbf{D}}^{\text{op}} \mid a'',M \rangle \langle a''M \mid V \mid a',M \rangle}{E_{a'} - E_{a''}} + \frac{\langle a,M \mid V \mid a'',M \rangle \langle a'',M \mid \vec{\mathbf{D}}^{\text{op}} \mid a',M \rangle}{E_{a} - E_{a''}} \right].$$
(3)

Because of the form of the operator  $\vec{D}^{op}$ , the sums over the intermediate states can be performed in an approximate way as follows.<sup>7</sup> Let us define

$$S(\vec{\xi}_{1},\vec{\xi}_{2}) = \sum_{a''} \frac{\langle a,M \mid V \mid a'',M \rangle \langle a'',M \mid \exp[-i(\vec{\xi}_{1} \cdot \vec{r}_{1} + \vec{\xi}_{2} \cdot \vec{r}_{2})] \mid a',M \rangle}{E_{a} - E_{a''}}, \tag{4a}$$

$$T(\vec{\xi}_{1}, \vec{\xi}_{2}) = \sum_{a''} \frac{\langle a, M \mid \exp[-i(\vec{\xi}_{1} \cdot \vec{r}_{1} + \vec{\xi}_{2} \cdot \vec{r}_{2})] \mid a'', M \rangle \langle a'', M \mid V \mid a', M \rangle}{E_{a'} - E_{a''}}, \tag{4b}$$

such that

$$\nu \langle a, M \mid \vec{\mathbf{D}}^{\text{op}} \mid a', M \rangle_{V} = -ie \left[ \left[ \frac{\partial}{\partial \vec{\xi}_{1}} + \frac{\partial}{\partial \vec{\xi}_{2}} \right] (S+T) \right]_{\vec{\xi}_{1} = \vec{\xi}_{2} = \vec{0}}. \tag{4c}$$

Let us also define the operators  $\hat{F}$  and  $\hat{G}$  such that

$$\{ [\hat{F}, H_0] + (E_a - E_{a'})\hat{F} \} | a' \rangle = \exp[-i(\vec{\xi}_1 \cdot \vec{r}_1 + \vec{\xi}_2 \cdot \vec{r}_2)] | a' \rangle , \qquad (5a)$$

$$\langle a \mid \{ -[\hat{G}, H_0] + (E_a - E_{a'}) \hat{G} = \langle a \mid \exp[-i(\vec{\xi}_1 \cdot \vec{r}_1 + \vec{\xi}_2 \cdot \vec{r}_2)] ,$$
 (5b)

where  $H_0$  is the unperturbed Hamiltonian of  $O_2$ . One then has  $S = \langle a, M \mid V\hat{F} \mid a', M \rangle$  and  $T = \langle a, M \mid \hat{G}V \mid a', M \rangle$ . To evaluate  $\hat{F}$  and  $\hat{G}$ , we assume that they depend only on position variables. This approximation permits us to replace  $H_0$  in (5a) and (5b) with the kinetic energy operator  $(\vec{p}_1^2/2m + \vec{p}_2^2/2m)$  of the valence electrons.  $\hat{F}$  and  $\hat{G}$  are then readily evaluated in the plane-wave representation, and S and T become

$$S = \sum_{\vec{k}_1, \vec{k}_2, \vec{k}_1', \vec{k}_2'} \frac{\phi_a^{\bullet}(\vec{k}_1, \vec{k}_2) \chi_a^{\dagger}(1, 2) \langle M \mid \mathcal{Y}(\vec{k}_1 - \vec{k}_1', \vec{k}_2 - \vec{k}_2') \mid M \rangle \chi_{a'}(1, 2) \phi_{a'}(\vec{k}_1' + \vec{\xi}_1, \vec{k}_2' + \vec{\xi}_2)}{E_a - E_{a'} - \sum_{j=1,2} \frac{(\vec{k}_j')^2}{2m} + \sum_{j=1,2} \frac{(\vec{k}_j' + \vec{\xi}_j)^2}{2m}},$$
 (6a)

$$T = \sum_{\vec{k}_{1}, \vec{k}_{2}, \vec{k}_{1}', \vec{k}_{2}'} \frac{\phi_{a}^{\bullet}(\vec{k}_{1} - \vec{\xi}_{1}, \vec{k}_{2} - \vec{\xi}_{2})\chi_{a}^{\dagger}(1, 2)\langle M \mid \mathcal{Y}(\vec{k}_{1} - \vec{k}_{1}', \vec{k}_{2} - \vec{k}_{2}') \mid M \rangle \chi_{a}(1, 2)\phi_{a}(\vec{k}_{1}', \vec{k}_{2}')}{E_{a} - E_{a} + \sum_{j=1,2} \frac{(\vec{k}_{j} - \vec{\xi}_{j})^{2}}{2m} - \sum_{j=1,2} \frac{(\vec{k}_{j})^{2}}{2m}}.$$
(6b)

Here  $\mathcal{V}(\vec{k}_1, \vec{k}_2)$  is the Fourier transform

$$\mathcal{Y}(\vec{\mathbf{k}}_{1},\vec{\mathbf{k}}_{2}) = \int d\vec{\mathbf{x}}_{1}d\vec{\mathbf{x}}_{2}e^{-i(\vec{\mathbf{k}}_{1}\cdot\vec{\mathbf{x}}_{1}+\vec{\mathbf{k}}_{2}\cdot\vec{\mathbf{x}}_{2})}V(\vec{\mathbf{x}}_{1},\vec{\mathbf{x}}_{2})$$

$$= \frac{10\pi g\mu_{B}^{2}}{3} \sum_{\lambda} \left[ \delta_{\vec{\mathbf{k}}_{2},\vec{0}}e^{-i\vec{\mathbf{k}}_{1}\cdot\vec{\mathbf{R}}_{\lambda}} \left[ \vec{\sigma}_{1}\cdot\vec{\sigma}_{\lambda} - 3\frac{(\vec{\sigma}_{1}\cdot\vec{\mathbf{k}}_{1})(\vec{\sigma}_{\lambda}\cdot\vec{\mathbf{k}}_{1})}{|\vec{\mathbf{k}}_{1}|^{2}} \right] + \delta_{\vec{\mathbf{k}}_{1},\vec{0}}e^{-i\vec{\mathbf{k}}_{2}\cdot\vec{\mathbf{R}}_{\lambda}} \left[ \vec{\sigma}_{2}\cdot\vec{\sigma}_{\lambda} - 3\frac{(\vec{\sigma}_{2}\cdot\vec{\mathbf{k}}_{2})(\vec{\sigma}_{\lambda}\cdot\vec{\mathbf{k}}_{2})}{|\vec{\mathbf{k}}_{2}|^{2}} \right] \right]. \tag{7a}$$

We also separated the  $O_2$  wave functions into spatial and spin parts as in  $\psi_a(\vec{x}_1, \vec{x}_2) \chi_a(1, 2)$ , and  $\phi_a$  is the Fourier transform of  $\psi_a$ :

$$\phi_a(\vec{k}_1, \vec{k}_2) = \int d\vec{x}_1 d\vec{x}_2 e^{-i(\vec{k}_1 \cdot \vec{x}_1 + \vec{k}_2 \cdot \vec{x}_2)} \psi_a(\vec{x}_1, \vec{x}_2) . \tag{7b}$$

It follows from (4c) that

737

$$\nu \langle a, M \mid \vec{\mathbf{D}}^{\text{op}} \mid a', M \rangle_{\nu} = -\frac{ie\vec{\pi}^{2}}{m (E_{a} - E_{a'})^{2}} \sum_{\vec{k}_{1}, \vec{k}_{2}, \vec{k}_{1}', \vec{k}_{2}'} \phi_{a}^{*}(\vec{k}_{1}', \vec{k}_{2}') \phi_{a'}(\vec{k}_{1}' - \vec{k}_{1}, \vec{k}_{2}' - \vec{k}_{2})(\vec{k}_{1} + \vec{k}_{2}) \times \left[\chi_{a}^{\dagger}(1, 2) \langle M \mid \mathscr{V}(\vec{k}_{1}, \vec{k}_{2}) \mid M \rangle \chi_{a'}(1, 2)\right]. \tag{8}$$

Now let a be the state  $^{1}\Delta_{g}$  with  $M_{L}=+2$ , and a' the state  $^{3}\Sigma_{g}^{-}$  with  $M_{S}=+1$ . The  $O_{2}$  wave functions for these states are given by  $^{11}$ 

$$\psi_a = \pi_e^+(\vec{\mathbf{x}}_1)\pi_e^+(\vec{\mathbf{x}}_2) , \qquad (9a)$$

$$\chi_{a} = \frac{1}{\sqrt{2}} \left[ \chi_{+}(1) \chi_{-}(2) - \chi_{-}(1) \chi_{+}(2) \right], \tag{9b}$$

$$\psi_{a'} = \frac{1}{\sqrt{2}} \left[ \pi_g^+(\vec{\mathbf{x}}_1) \pi_g^-(\vec{\mathbf{x}}_2) - \pi_g^-(\vec{\mathbf{x}}_1) \pi_g^+(\vec{\mathbf{x}}_2) \right], \tag{9c}$$

$$\chi_{a'} = \chi_{+}(1)\chi_{+}(2)$$
, (9d)

where  $\pi_g^{\pm}(\vec{x})$  are molecular orbitals for individual electrons, and  $\chi_{\pm}$  are the electronic spinors for spin up and down. A straightforward algebra then yields

$$\vec{\mathbf{D}} = {}_{V} \langle {}^{1}\Delta_{\mathbf{g}}, M_{L} = 2; M \mid \vec{\mathbf{D}}^{\text{ op}} | {}^{3}\Sigma_{\mathbf{g}}^{-}M_{S} = 1; M \rangle_{V}$$

$$= \frac{i75eg\mu_B^2 \tilde{\pi}^2}{32\pi m (\Delta E)^2} \sum_{\lambda} \langle M \mid \sigma_{\lambda z} \mid M \rangle \frac{\partial}{\partial \vec{R}_{\lambda}} \left[ \int d\vec{x} [\pi_g^+(\vec{x})]^{\bullet} \frac{\hat{\epsilon}_+ \cdot (\vec{x} - \vec{R}_{\lambda})}{|\vec{x} - \vec{R}_{\lambda}|^4} \pi_g^-(\vec{x}) \right], \tag{10}$$

where  $\Delta E = E_a - E_{a'}$  is the unperturbed energy difference and  $\hat{\epsilon}_{\pm}$  are the complex vectors  $\hat{\epsilon}_{\pm} = \hat{x} \pm i \hat{y}$ . The matrix element  $\langle M \mid \sigma_{\lambda z} \mid M \rangle = s_{\lambda}$ .  $\pi_g^{\pm}(\vec{x})$  can be written in terms of the n=2, l=1,  $m=\pm 1$  atomic orbitals  $p_{\pm}(\vec{x})$  as

$$\pi_g^{\pm}(\vec{x}) = p_{+}(\vec{x} - \vec{a}) - p_{+}(\vec{x} - \vec{b}),$$
 (11)

where  $\vec{a}$  and  $\vec{b}$  are the positions of the oxygen atoms. To obtain an estimate for (10), we first simplify (11) by approximating  $\pi_{\vec{e}}^{\pm}$  with

$$\pi_{\mathbf{g}}^{\pm}(\vec{\mathbf{x}}) \simeq -\vec{\mathbf{r}}_{ab} \cdot \vec{\nabla} p_{\pm}(\vec{\mathbf{x}}) ,$$
 (12a)

where  $\vec{r}_{ab} = \vec{a} - \vec{b}$  is the internuclear distance of  $O_2$ . Second, we assume that external spins are relatively far from  $O_2$  and use the expansion

$$\frac{1}{|\vec{x} - \vec{R}|^4} \simeq \frac{1}{R^4} + \frac{4(\vec{x} \cdot \vec{R})}{R^6} + \frac{[12(\vec{R} \cdot \vec{x})^2 - R^2 x^2]}{R^8}.$$

Using Eqs. (12a) and (12b), the integral in Eq. (10) can be readily evaluated. The result is somewhat long; however, it can be simplified by taking an average over the orientations of the molecular axis (i.e., over  $\hat{r}_{ab}$ ), and also over the orientations of the external spins (i.e., over  $\hat{R}_{\lambda}$ ), if N is sufficiently large and the spins are distributed around the molecule. One then finds

$$\langle\langle \vec{\mathbf{D}} \rangle\rangle \simeq \frac{i75 eg \mu_B^2 \hbar^2 r_{ab}^2}{14\pi m (\Delta E)^2} (3\hat{e}_- + \hat{z}) \sum_{\lambda} s_{\lambda} \frac{1}{R_{\lambda}^6} . \tag{13}$$

The sum in Eq. (13) can be replaced by an expression which involves the probabilities of finding the spin up

 $(P_{+})$  or down  $(P_{-})$  at a given site, and a structure factor for the external spin system,

$$\sum_{1} \frac{s_{\lambda}}{R_{1}^{6}} \rightarrow (P_{+} - P_{-}) \sum_{\lambda} \frac{1}{R_{1}^{6}} \equiv (P_{+} - P_{-}) \frac{1}{r_{0}^{6}} Z, \quad (14)$$

where  $r_0$  is a scaling length such that  $r_0 = 10^{-8}$  cm. Z may be calculated in a similar manner to the calculation of a Madelung constant. For an order of magnitude determination, however, one may replace the sum over  $\lambda$  by a three-dimensional volume integral, and Z is approximately given by

$$Z = \sum_{\lambda} \left[ \frac{r_0}{R_{\lambda}} \right]^6 \approx \frac{4\pi}{3} \left[ \frac{r_0}{r_s} \right]^3, \tag{15}$$

where  $r_s$  is the average distance between two spin sites (hence  $r_s$  is the smallest length in the set  $\{R_{\lambda}\}$ ). For many magnetic materials  $r_s/r_0 \sim 2.9$  Setting g=2,  $\Delta E=1$ , eV, and  $r_{ab}/r_0 \sim 1.5$ , we find

$$\langle\langle \vec{\mathbf{D}} \rangle\rangle \approx (3 \times 10^{-3} \text{ debye})i(3\hat{\epsilon}_{\perp} + \hat{z})(P_{\perp} - P_{\perp})$$
. (16)

In other words, the induced moment is on the order of  $10^{-2}$  debye, and the induced spontaneous decay rate is on the order of  $10^2 \text{ sec}^{-1}$ . Furthermore, if the perturbed  $O_2$  is in a laser field of power p, then its Rabi frequency is

$$\Omega_R = \frac{\langle \langle \vec{\mathbf{D}} \rangle \rangle \cdot \vec{\mathbf{E}}}{\hbar} \sim \frac{D}{\hbar} \left[ \frac{4\pi p}{c} \right]^{1/2} \sim 10^9 \text{ sec}^{-1} , \qquad (17)$$

for  $p = 10^7 \text{ W/cm}^2$ .

<u> 29</u>

#### III. CONCLUDING REMARKS

It is clear from the preceding discussion that if the external spins have no preferred direction and are completely random, the induced dipole moment vanishes within the approximations made above. In order to obtain dipole moment between  ${}^{1}\Delta_{g}$  and  ${}^{3}\Sigma_{g}^{-}$  states, the singlet oxygen must interact with a collection of particles which has net magnetization over some finite volume.

The best way to establish contact between the singlet oxygen and a magnetic surface may be by means of grazing-angle scattering. We have in mind the situation in which slowly moving  $(v \sim 10^3 - 10^5 \text{ cm/sec})$  singlet oxygen molecules scatter from a ferromagnetic surface at grazing angles (<1°), and the stimulated emission occurs while molecules are in the vicinity of the surface. This type of scattering permits maximum exposure of the metastable molecules to the localized spins of the magnetic material, and minimizes the damage to the surface. The same surface can therefore be used repeatedly. <sup>10</sup>

An important advantage of the above scattering scheme is that it cuts down on possible chemical reactions between the singlet oxygen and the surface by restricting the duration of the contact. In chemical reactions between the singlet oxygen and the ferromagnetic materials, the quenching rate  $k_Q$  is typically  $10^8 \text{ mol}^{-1} \sec^{-1}$ . Let us consider a single  $O_2$  molecule and assume that it follows a straight line trajectory on the surface with a constant velocity v for a distance l. Let  $\sigma_0$  be the effective cross-sectional area of this molecule. The number of effective moles of the material atoms that it sees on this trajectory

is  $\sigma_0 l \rho_M / N_A$ , where  $\rho_M$  is the number of atoms per unit volume of the target material and  $N_A$  is the Avogadro's number. Thus the overall transition rate is  $k_Q \sigma_0 l \rho_M / N_A$ . Since the duration of the interaction is l/v, the probability of quenching is given by

$$pr_Q \sim \frac{k_Q \sigma_0 l^2 \rho_M}{N_A v} \tag{18}$$

on the basis of this simple model. Taking  $N_A = 6 \times 10^{23}$ , l = 1 cm,  $\sigma_0 \sim 10^{-14}$  cm<sup>2</sup>,  $\rho_M \sim 10^{22}$  atom/cm<sup>3</sup>, and  $v \sim 10^3$  cm/sec, one finds that  $pr_Q \sim 10^{-11}$ , which rules out chemical quenching.

There are, of course, other types of quenching mechanisms, such as electron pick-up or electron loss by the scattering molecule, exchange of the molecular rotational energy with the surface, etc. Furthermore, the surface may partially be demagnetized as a result of the scattering. However, we do not expect these processes to alter the order of magnitude results presented above. We conclude that the scheme proposed here appears to be quite promising to obtain an oxygen laser. More importantly, the scheme may be used in obtaining lasers from other metastable species for which collision partners are not known at the present time.

### **ACKNOWLEDGMENTS**

This research was supported by U. S. Air Force Weapons Laboratory, Kirtland Air Force Base, and the the U. S. Air Force Office of Scientific Research.

<sup>&</sup>lt;sup>1</sup>M. Kasha and D. E. Brabham, in Singlet Oxygen, Organic Chemistry, edited by R. W. Murray (Academic, New York, 1979), Vol. 40.

<sup>&</sup>lt;sup>2</sup>R. M. Badger, A. C. Wright, and R. F. Whitlock, J. Chem. Phys. <u>43</u>, 4345 (1965).

<sup>&</sup>lt;sup>3</sup>V. S. Zuev, V. A. Katulin, V. Yu. Nosach, and O. Yu. Nosach, Zh. Eksp. Teor. Fiz. <u>62</u>, 1673 (1972) [Sov. Phys.—JETP <u>35</u>, 870 (1972).

<sup>&</sup>lt;sup>4</sup>A. Elci and D. Rogovin, Phys. Rev. Lett. <u>48</u>, 864 (1982).

<sup>5</sup>K. Druhl, M. O. Scully, and A. W. Overhauser, Opt. Commun. 38, 393 (1981).

<sup>&</sup>lt;sup>6</sup>D. Rogovin and P. Avizonis, Appl. Phys. Lett. <u>38</u>, 666 (1981).

<sup>&</sup>lt;sup>7</sup>A. Dalgarno and J. T. Lewis, Proc. R. Soc. London Ser. A <u>233</u>, 70 (1955).

<sup>&</sup>lt;sup>8</sup>M. Kotani, Y. Mizuno, K. Kayama, and E. Ishiguro, J. Phys. Soc. Jpn. <u>12</u>, 707 (1957).

<sup>&</sup>lt;sup>9</sup>C. Kittel, Introduction to Solid State Physics (Wiley, New York, 1971).

<sup>&</sup>lt;sup>10</sup>C. Rau and R. Sizmann, in *Atomic Collisions in Solids*, edited by S. Datz, B. R. Appleton, and C. D. Moak (Plenum, New York, 1975), Vol. 1.

<sup>&</sup>lt;sup>11</sup>D. Bellus, in Singlet Oxygen, edited by B. Ranby and J. F. Rabek (Wiley, New York, 1978).

field are small. When this is the case, Eqs. (1)-(3) may be linearized to obtain

$$\partial K_2(z,\tau_0,\mu_0)/\partial z = K_1(z,\tau_0,\mu_0)$$
, (7)

$$\partial K_1(z,\tau_0,\mu_0)/\partial z = \kappa A_i^* e E_S(z,\tau) , \qquad (8)$$

$$\partial E_{s}(z,\tau)/\partial z = iD A_{i} \int d\mu_{0} \mathcal{J}(\tau_{0},\mu_{0}) e^{-i\mu_{0}z} K_{2}(z,\tau_{0},\mu_{0})$$
 (9)

 $K_2 = -(1/2\pi) \left[ d\theta_0 \exp(-i\theta_0) \delta\theta \right]$  and  $K_1 = -(1/2\pi) \left[ d\theta_0 \exp(-i\theta_0) (\mu - \mu_0) \right]$  are respectively the density and energy bunching amplitudes. We expect the FEL to saturate when  $|K_2|$  becomes comparable to one. The initial values of  $K_2$  and  $K_1$  are zero.

If there is no inhomogeneous broadening, we set  $\mathcal{J}(\tau_0,\mu_0)=I(\tau_0)^\delta(\mu_0)$ . Then Eqs. (7)-(9) may be solved by Laplace transforming in z to give

$$\mathsf{E}_{\mathsf{S}}(z,\tau) = \mathsf{E}_{\mathsf{S}}(0,\tau)$$

+ 
$$\int_{0}^{z} dz' \frac{1}{z'} F_0(x) E_s(0, \tau - z'/2\gamma_s^2 c)$$
, (10)

where  $F_0\left(x\right)$  is a generalized hypergeometric function given by the series expansion

$$F_0(x) = \sum_{n=1}^{\infty} \frac{1}{n!(2n-1)!} x^n$$
 (11)

and

$$x = i\kappa D[A_i]^2 2\gamma_s^2 cz'^2 \int d\tau_0 I(\tau_0) .$$

$$- \tau - z/2\gamma_s^2 c$$
(12)

Equation (10) expresses the field at position z as the sum of the incident field at time  $\tau$  and an integral over the incident field at earlier times. We see explicitly that coherence cannot develop in one pass over more than the slippage time  $L/2\gamma_S^2c$ , where L is the wiggler length. This is true also in the strong-signal regime. In other words, an FEL operating without mirrors by ASE cannot be coherent over more laser periods than there are wiggler periods.

In the case that both the current and the laser field are cw we put  $E_s(z,\tau) = E_s(z) \exp(i2\gamma_s^2 c \tilde{\omega}_0 \tau)$  in Eq. (10) to obtain

$$E_{s}(z) = E_{s}(0) \left[1 + \int_{0}^{z} dz' \frac{1}{z'} F_{0}(iCz'^{2}(z - z'))e^{-i\overline{\mu_{0}}z'}\right], \quad (13)$$

where  $C = \kappa D|A_i|^2 I$ . Since the power series (11) is very rapidly convergent, the field can generally be calculated accurately by keeping only a few terms in the expansion. Only the first term is needed to obtain the well-known antisymmetric small-gain formula with peak gain .27  $CL^3$  at  $\overline{\nu_0}L = 2.6$ .

An alternative approach to cw theory is to drop the dependence on  $\tau$  and  $\tau_0$  in Eqs. (1)-(3) and (7)-(9). In this case we take  $\mathcal{J}(\mu_0)$  to be centered at  $\mu_0$  with width U. That is, the detuning  $\overline{\mu}_0$  from resonance is included in  $\mathcal{J}$  rather than  $E_S$ . If we define the normalized distribution  $f(\mu_0)$  by  $\mathcal{J}=$  If, then Eqs. (7)-(9) become

$$dK_2(z,\mu_0)/dz = K_1$$
, (14)

$$dK_1(z,\mu_0)/dz = \kappa A_i^* e^{i\mu_0 z} E_s$$
, (15)

$$dE_{s}(z)/dz = iD A_{i}I \int d\mu_{0}f(\mu_{0})e^{-i\mu_{0}z} K_{2}(z,\mu_{0}) . \qquad (16)$$

There equations may be solved by Laplace transforming to give

$$E_{s}(z) = \frac{1}{2\pi i} E_{s}(0) \int_{a-i\infty}^{a+i\infty} d\beta e^{\beta z} [\beta - iC \int d\mu_{0} f(\mu_{0})/(\beta + i\mu_{0})^{2}]^{-1}, \quad (17)$$

where the contour is to be taken to the right of all singularities. The simplest case is where  $f(\mu_0)$  is a Lorentzian,

$$f(\mu_0) = \frac{1}{\pi U} \frac{1}{1 + \left(\frac{\mu_0 - \bar{\mu}_0}{U}\right)^2} . \tag{18}$$

Then the integral over  $\mu_0$  in Eq. (17) gives  $(\beta + i \overline{\mu_0} + U)^{-2}$ . We may close the contour integral over  $\beta$  around the three poles at the roots  $\beta$ , of the equation

$$\beta(\beta + i\overline{\mu_0} + U)^2 = iC \tag{19}$$

and evaluate the integral to express  $E_s(z)$  as a linear combination of the three exponentials  $\exp(\beta_1 z)$ . In the case U = 0 it can be shown that this solution is equivalent to that in Eq. (13). In general at most one of the three roots has a positive real part, and the corresponding mode will dominate near the end of a sufficiently long wiggler (unless saturation sets in first). The detuning giving maximum growth of the unstable mode is  $\overline{\mu_0} = U/3^{\frac{1}{2}}$ . The gain is greatly reduced if  $U \gtrsim C^{\frac{1}{3}}$ . In the case that  $U < C^{\frac{1}{3}}$  and  $\overline{\mu_0} = U/3^{\frac{1}{2}}$  the three modes contribute about equally at the entrance of the wiggler and the gain in the limit of very large gain approaches

$$G = \frac{1}{9} \exp \left[ \left( 3^{1/2} C^{1/3} - \frac{4}{3} U \right) L \right] . \tag{20}$$

Graphs showing the dependence of  $2\text{Re}(\beta)$  on  $\mu_0$  and U may be found in Reference 4. The condition that the FEL saturates when  $K_2=1$  can be used to infer a saturation power  $P_{sat}=(I^2A_1/4k_q)^{73}(c/\epsilon_0\Sigma)^{1/3}$ .

## SEMICLASSICAL FEL GAIN

When electrons emit a laser photon, they undergo a recoil  $\delta E = \hbar \, \omega_S$ . If we let the recoil in energy detuning units be 2q, we see from Eq. (4) that

$$q = \hbar k_s^2 / 2Mc\gamma_s^3 . (21)$$

If 2q is comparable to or larger than the homogeneous broadening  $\pi/L$ , then the classical gain formula becomes incorrect and seriously overestimates the true gain. The quantum recoil can be important for an electromagnetic wiggler at infrared wavelengths. In this case  $\gamma$  is typically  $\stackrel{<}{\sim}$  50 and  $\overline{\Delta}=1$  is small. Note that L for an electromagnetic wiggler is only half the wiggler pulse length (L =  $\frac{1}{2}$  cTp), since the wiggler pulse and the electrons pass through each other. The product 2qL may be written as

$$2qL = 582\pi \frac{T_{p}(nsec)}{[\lambda_{i}(\mu m)]^{\frac{3}{2}}[\lambda_{s}(\lambda)]^{\frac{1}{2}}}.$$
(22)

As an example, for a CO<sub>2</sub> laser pulse 1 nsec long used to generate radiation of  $\lambda_s$  = 100Å, Eq. (22) gives 1.6 $\pi$ .

It has been shown by Renieri<sup>5</sup> that the condition  $2qL > \pi$  implies that the quantum-mechanical spreading of the wave packet describing the electron is larger than the laser wavelength. This is another indication that a quantum-mechanical treatment of the electrons is required.

We present here a semiclassical theory of the FEL in which the electrons are described quantum mechanically, while the field is treated classically. This approach accounts satisfactorily for the quantum recoil but (as in Lamb's semiclassical theory of the laser one include the noise needed to produce laser start-up when no laser field is initially present. In the laboratory frame the semiclassical theory can be developed by coupling the Klein-Gordon equation for the electrons with the Maxwell equation for the field. An equivalent approach is obtained by recalling that  $\theta$  and  $\mu$  are proportional to the position and momentum of an electron in a frame moving at the speed of the ponderomotive potential. In quantum theory these become operators  $\hat{\theta}$  and  $\hat{\mu}$  whose commutator is

$$[\hat{\theta}, \hat{\mu}] = 2iq . \tag{23}$$

Restricting our attention to cw operation, Eqs. (1) and (2) become the operator equations

$$d\hat{\theta}/dz = \hat{\mu}, \tag{24}$$

$$\hat{d\mu}/dz = -\kappa [A_i^* E_s(z)e^{i\hat{\theta}} + c.c.] . \qquad (25)$$

We consider here only the case of a uniform wiggler, so A; is constant. For a wiggler field which is a high-power Gaussian beam, the wiggler is effectively tapered by the slowing of the electrons in the vicinity of the beam waist. This is an important source of additional homogeneous broadening if the pump laser power exceeds  $\pi \epsilon_0 \, m^2 c^5/2e^2 \approx 1 \, \text{GW}$ .

. Equations (24) and (25) may be interpreted as Heisenberg equations of the "Hamiltonian"

$$H = \frac{1}{2} \hat{\mu}^2 - i \kappa A_i^* E_s e^{i\hat{\theta}} + i \kappa A_i E_s^* e^{-i\hat{\theta}}$$
 (26)

with the following rule for the evolution of operators:

$$\partial \hat{A}/\partial z = (i/2q)[H, \hat{A}]$$
 (27)

Similarly the Schrödinger equation for state vectors becomes

$$H|\psi\rangle = 2iqd|\psi\rangle/dz . \qquad (28)$$

This formulation is equivalent to nonrelativistic quantum mechanics in the moving frame. The semiclassical generalization of Eq. (3) is obtained by replacing the averages on the right side by the quantum expectation value, yielding

$$dE_{s}(z)/dz = DA_{i}I tr(\rho e^{-i\hat{\theta}}), \qquad (29)$$

where  $\rho$  is the density matrix. If we assume that the initial density matrix is diagonal in the  $\mu$  representation,

$$\rho(\mu_0, \mu_0^*) = f(\mu_0)\delta(\mu_0 - \mu_0^*) , \qquad (30)$$

we can use first-order perturbation theory to obtain a quantum generalization of Eqs. (14)-(16). It turns out that only Eq. (15) is altered. It becomes

$$dK_1(z,\mu_0)/dz = \kappa A_i^* e^{i\mu_0 z} E_s - q^2 K_2.$$
 (31)

If  $f(\mu_0)$  is Lorentzian, the cubic dispersion relation (19) is generalized to become

$$\beta[(\beta + i\overline{\mu}_0 + U)^2 + q^2] = iC.$$
 (32)

We show in Reference 4 that the quantum recoil acts to reduce  $2Re(\beta)$  for the unstable mode below its classical value, as well as to reduce the range in  $\overline{\mu}_0$  over which there is an instability.

For the case of small gain per pass, the gain can be written quite generally as

$$G = C \frac{1}{2q} \int d\mu_0 [\dot{f}(\mu_0 + q) - f(\mu_0 - q)] \left( \frac{\sin \mu_0 L/2}{\mu_0/2} \right)^2.$$
 (33)

If 2q is very small compared to either the homogeneous broadening  $\pi/L$  or the inhomogeneous broadening U, then G reduces to its classical value. The condition U < 2q is rather restrictive; it is equivalent to the condition that the energy spread of the electrons be less than  $\hbar\omega_S$ . For 100Å x rays this is less than 1 keV. Even if low energy electrons are used (as would be the case with an optical wiggler), the relative energy spread needed to produce quantum modifications in the gain would still be less then .01%. However, even in the regime U > 2q, if  $2q > \pi/L$ , quantum effects should be manifested, for example, in the statistics of the emitted photons or in the electron energy distribution at saturation. When  $2q > \pi/L$ , one expects the FEL to saturate when one photon has been emitted per electron.

The validity of first-order perturbation theory depends on the smallness of the product of the interaction energy with the interaction time divided by  $\hbar$ . In terms of the Hamiltonian (26) one requires  $\kappa |A_i E_S| L < 2q$ . Substituting for the values of  $\kappa$  and q, this becomes (e²  $|A_i E_S|/2Mc^2\gamma_S\hbar k_S)L < 1$ . For an x-ray FEL (in contrast to, say,an infrared FEL) this condition will hold for relatively large laser intensities ( $E_S$  of the order of  $10^6 V/m$  with an optical wiggler), making first-order perturbation theory appropriate to deal with at least the small-signal regime. In the classical regime (2qL  $<<\pi$ ) it turns out that the gain predicted by perturbation theory is correct even for conditions where perturbation theory would be expected to break down, but it remains to be seen whether this is true of the quantum regime.

Further discussion of the quantum regime for an FEL with an infrared laser pulse as the wiggler may be found in Reference 4, where numerical examples are given for which the gain per pass may be as high as 40% at 5.7Å.

# CONSTRAINTS ON A LONG-WIGGLER X-RAY FEL

One can anticipate that the extension of FEL operation to x-ray wavelengths by using high-energy electrons and a long conventional wiggler will be a gradual and evolutionary one, in which one builds on the experience gained by previous experiments at longer wavelengths. Electron beam quality appears to be the greatest problem, although there are also difficult problems in resonator optics and wiggler construction. For soft x-rays storage rings provide an acceptable beam quality, and experiments using a storage ring to drive a 100Å FEL are being planned. Below about 60Å the storage-ring approach becomes untenable because of the quantum energy spread in the electron beam induced by synchrotron radiation in the ring. In principle one could get a better quality beam at high energy by using a linear accelerator, but suitable accelerators have not yet been built. It appears that rf linacs give too much energy spread, but induction linacs may offer a possibility. 10

We now list several constraints on the operation of a high-gain FEL, referring to a numerical example from Reference 4, the parameters of which are summarized in Table 1. Many of these constraints may be expressed as limitations on the length L for which a cold-beam classical one-dimensional gain calculation is correct.

Table I Parameters for a 5 Å X-Ray FEL

Quantity	Symbol	<u>Value</u>
Wiggler wavelength	$\lambda_{\mathbf{q}}$	3.2 cm .
Magnetic field	$\frac{\vec{\mathbf{B}}}{\Delta}$	.24 T
Mass shift	$\overline{\Delta}$	1.512
Laser wavelength	$\lambda_{S}$	5 Å
Electron energy	E	3.554 GeV
Current	I	10 A
Laser mode area	Σ	.16 mm <sup>2</sup>
Wiggler length	L	276 m
Gain	G	1000
Saturation power	$P_{Sat}$	1.7 NW
Slippage time	L/2Ys2c	$1.44 \times 10^{-14} sec$
Characteristic gain length	3-1/3C -1/3	30.29 m
Filling factor		1 .
Electrons per laser wavelength	Iλ <sub>S</sub> /ec	104

A. Reciprocal quantum width:  $\pi/2q = \pi Mc\gamma_s^3/\hbar k_s^2 = 11.5 \text{ km}$ . Since this is  $\gg$  L, the device is completely classical.

B. Diffractive spreading of laser beam: Rayleigh range =  $\Sigma/\lambda_q$  = 320 m. This is of the same order as L, so diffraction is a significant effect.

C. Reciprocal energy spread:  $1/U = (\lambda_q/4\pi) (E/\delta E)$ . If we take the energy spread  $\delta E$  to be 10 keV we obtain 1/U = 900 m, which is larger than L. Equation (20) indicates that the gain is reduced from 1000 to 664. The difficulty is whether one can obtain such a small energy spread at this high energy. This is presently unclear.

D. Wiggle amplitude: One must check that the wiggle amplitude does not exceed the assumed cross-sectional dimension of the beam. In the present case the wiggle amplitude is  $eB\lambda^2c/4\pi^2E$  = .5 µm, so there is no problem.

E. The characteristic distance over which the electron beam spreads due to Coulomb repulsion 11 is  $(4\epsilon_0\,\text{mc}^3\gamma^3\Sigma/\text{Ie})^{\frac{1}{2}}=5.4\,\text{km}$ . This is clearly no problem.

F. The characteristic distance over which the electron beam spreads due to emittance  $\varepsilon$  is  $\Sigma/\pi\varepsilon$ . If we assume that the normalized emittance  $\varepsilon_n$  is the same as for the Stanford superconducting linac, then  $\varepsilon$  = .06 mm mrad (43MeV/3.554 GeV). Then  $\Sigma/\pi\varepsilon$  = 70 m. This is small enough to be a problem. However, it is a problem which should be correctable by placing quadrupole focusing elements along the wiggler.

G. The condition that electrons with emittance stay in phase with the ponderomotive potential imposes a constraint on the length of the wiggler. The phase angle change for electrons of velocity component v is

$$\delta\theta = \int_{0}^{L} dz \left[ k_{q} - k_{s} (c/v_{z} - 1) \right]$$

$$= \int_{0}^{L} \left[ k_{q} - (k_{s}/2\gamma^{2})(1 + |P - eA_{z}|^{2}/m^{2}c^{2}) \right], \qquad (34)$$

where P is the transverse canonical momentum.

Since  $k_q - (k_s/2\gamma^2)(1 + e^2A_i^2/m^2c^2) = k_q - k_s/2\gamma_s^2 = 0$ , only the terms  $P \cdot A_i$  and  $P^2$  survive in the integrand. However,  $P \cdot A_i$  is oscillatory and averages to zero. Therefore

$$|\delta\theta| \approx (k_s/2\gamma^2)(P^2/m^2c^2)L \approx (k_q P^2/m^2c^2)L$$
 (35)

We may relate P to the emittance by

$$P^2 = (\epsilon^2/\Sigma)p_z^2 = (\epsilon^2/\Sigma)m^2c^2\gamma^2 = m^2c^2\epsilon_n^2/\Sigma$$
,

so that if we require  $|\delta\theta|<2\pi$ , we get  $L<\Sigma\lambda_q/\epsilon_n^2=201$  m. This is small enough to be significant.

H. Transverse dependence of the wiggler field leads to dephasing of electrons which sample the field at different transverse positions r. Let us assume that the wiggler field varies quadratically off axis, such that  $\delta A/A \simeq r^2/\lambda_q^2$ . The actual electron motion is rather complicated (betatron oscillations), but we can get a rough estimate by letting  $A_i$  vary with r in Eq. (34) (now setting P = 0). This gives  $\left|\delta\theta\right| \simeq k_q L \delta (e^2 \left|A_i^2\right|^2/m^2c^2)$ . If we assume that  $e^2 \left|A_i\right|^2/m^2c^2$  is of the order of unity, which is typically the case for magnetostatic wigglers used in FEL's, and let the variation in  $r^2$  be  $\Sigma$ , then  $\left|\delta\theta\right| \simeq k_q L \Sigma/\lambda_q^2$ . The condition  $\left|\delta\theta\right| < 2\pi$  now becomes L  $<\lambda_q^3/\Sigma$  = 205 m. This is small enough to be a problem. Note that the length constraint here is proportional to  $1/\Sigma$ , in contrast to the constraints in items B, E, F and G, which are proportional to  $\Sigma$ .

I. To operate without a resonator by ASE, there must be sufficient noise to get the lasing started. For conventional lasers there is a characteristic delay time after which the lasing starts. For the FEL there is an analogous delay distance. According to Bonifacio, Narducci and Pellegrini,  $^{12}$  the delay distance  $z_{\rm D}$  is given by the product of the characteristic gainlength  $3^{-1/2} {\rm C}^{-1/3}$  and  $(\ln |N| + 1)$ , where N is the number of electrons per laser wavelength. This criterion gives  $z_{\rm D}$  = 10lm, which means that lasing should occur without a resonator, at least on most shots.

A disadvantage of operating without a resonator is that the temporal coherence is limited to the slippage time, which is only 1.44 x  $10^{-14}$ sec for the present example. Perhaps an order of magnitude increase in temporal coherence can be obtained by using a Bragg crystal resonator, simply because of the narrow wavelength band  $\delta\lambda$  over which the resonator is a good reflector. This will extend the optical wave packets to a minimum length  $\lambda_S^2/c\delta\lambda$  after reflection. Nevertheless, it is unlikely that an x-ray FEL can approach the coherence obtainable by free-electron harmonic up-conversion of an already coherent laser signal.

In conclusion, it appears that extension of FEL operation down to the 5 Å range will be difficult, but maybe not impossible. It will depend on experience gained by doing high-gain FEL experiments at longer wavelengths and on developments in accelerator technology motivated by FEL applications. An FEL with a long conventional wiggler is capable of higher gain than one using an infrared electromagnetic wiggler. On the other hand, the latter type of device is more compact, uses low-energy electrons, and presents an opportunity to explore the quantum regime of the FEL.

## **ACKNOWLEDGMENTS**

This research was supported by the Office of Naval Research and the Air Force Office of Scientific Research.

# REFERENCES

- 1. G. T. Moore and M. O. Scully, Phys. Rev. A21 2000 (1980).
- 2. H. Al-Abawi, G. T. Moore and M. O. Scully, Phys. Rev. A24, 3143 (1981).
- 3. R. Bonifacio, P. Meystre, G. T. Moore and M. O. Scully, Phys. Rev. A21, 2009 (1980).
- 4. J. Gea-Banacloche, G. T. Moore and M. O. Scully, "Prospects for an X-Ray Free-Electron Laser," proceedings of the Orcas Island FEL Conference, to be published by SPIE.
- 5. A. Renieri, see his paper in this volume.
- 6. W. E. Lamb, Jr., Phys. Rev. 134, A1429 (1964).
- 7. M. V. Fedorov, IEEE J. Quantum Elec. QE-17, 1359 (1981).
- 8. J. M. J. Madey, talk at Stanford New Rings Workshop, July, 1983. Also see his paper in this volume.
- A. Renieri, "The Free-Electron Laser: The Storage Ring Operation", in Developments in High-Power Lasers and Their Applications, ed. C. Pellegrini, (North Holland, 1981), p. 431.
- 10. J. M. J. Madey, private communication.
- 11. J. D. Lawson, The Physics of Charged-Particle Beams (Oxford University Press, 1977), p. 136.
- 12. R. Bonifacio, L. Narducci and C. Pellegrini, see their paper in this volume.
- 13. R. Colella and A. Luccio, see their paper in this volume.

# APPENDIX E

# FORBIDDEN NUCLEAR BETA-DECAY IN AN INTENSE PLANE WAVE FIELD

W. Becker\*, R. R. Schlicher and M. O. Scully\* +

Institute for Modern Optics
Department of Physics and Astronomy
University of New Mexico
Albuquerque, New Mexico 87131

Max-Planck-Institut für Quantenoptik
D-8046 Garching bei München
West Germany

## Abstract

We present the nonrelativistic theory of nuclear beta-decay of arbitrary forbiddenness in the presence of an external plane wave field. Emphasis is on the question of whether the field is able to break the common selection rules, so that forbidden decays could be significantly enhanced. It turns out that while this is true in principle the required field strength is of the order of the critical one which is far beyond experimental possibilities. Recent claims to the contrary are contested. In particular, in the case of the first order forbidden decay of <sup>90</sup>Sr, we obtain an enhancement which is smaller than a previously published estimate by twenty-four orders of magnitude.

# I. Introduction

Nuclear decay rates are usually considered to be, on earth and under even extreme laboratory conditions, almost on the same footing with fundamental constants. For example, they are commonly supposed to be essentially independent of changes in their environment as specified by temperature, external electromagnetic fields, etc.

Recently, however, it has been claimed [1,2] that total decay rates of forbidden nuclear beta-decay can be significantly enhanced by an intense plane wave field. The idea was that external fields be able to change the selection rules of the decay or, at least, to generate contributions to the decay rate which are of the same type and order of magnitude as those which usually allow for forbidden decays. Whereas it is beyond any doubts that this can be achieved with fields of the order of the critical field strength  $E_{crit} = (m^2c^3/e^{\frac{1}{2}})$  $\simeq 1.3 \times 10^{16}$  Volt/cm, the claim is that experimentally accessible fields are already sufficient in order to render the effect observable, e.g., focused intense laser fields with a field strength of about 10 Volt/cm or radio fields of a transmission line with even much smaller field strengths. The original estimates [2] suggesting enhancements by many orders of magnitude for highly forbidden decays were shown to be erroneous [3]. However, in Refs. 4 and 5, significant enhancements are still predicted. The present authors contested these statements on the basis of general arguments in Ref. 3. However, neither in Ref. 3, nor in Refs. 4 and 5 were quantitative estimates of the total decay rate given.

In this paper, we derive and evaluate explicitly the total rate for a decay of arbitrary forbiddenness in the presence of an intense plane wave field. Our theoretical model will be essentially that of Refs. 1 and 2. The electron will be described by the Volkov solution for a charged particle in the presence of a

plane wave field. The nuclear model will be the shell model with an inert of-core, which is affected neither by the decay nor by the external field, and one or more valence nucleon in an angular momentum coupled state, so that the nuclear wave functions are specified by an effective one-particle Hamiltonian. However, as will be pointed out later, we disagree with Refs. 1 and 2 on the interpretation of the nuclear wave functions in the external field. We shall achieve great formal simplification by restricting ourselves to a monrelativistic treatment in the spirit of Fermi's fundamental paper [6]. Hence, our results are not immediately applicable to real decays. However, the purpose of this note is to give simple and explicit results for an albeit idealized situation, which, if positive, would encourage a more realistic treatment. Moreover, we have initiated an analogous relativistic treatment [3], the conclusion being that the field dependence is much the same in the relativistic and nonrelativistic case.

It should be mentioned, that we shall entirely disregard effects which are due to the atomic environment. It is well known that external parameters such as temperature do affect observed nuclear decay rates via their impact on the electron's wave function, in particular, if it is bound, like in electron capture, bound state beta-decay or internal conversion. These effects have been estimated and been found to be of relative order of 10<sup>-2</sup>, at best, under favorable conditions; for a review see [7], see also [8]. In contrast, the effects predicted in [1,2] are much larger.

In Section II we shall set up the general framework. In contrast to Refs. 1, 2 and 4, 5 we shall directly calculate the total decay rate. This means, we first square the S-matrix element for the decay, then integrate over the final phase space, and do the integrations over time last. This procedure which has previously been found to be advantageous in external field problems (see, e.g.,

Ref. 9) leaves the dependence on the external field transparent throughout the calculation and bypasses the cumbersome and error prone integration over the differential decay rate. In Section III we proceed to an explicit evaluation of the total decay rate for a monochromatic plane wave field with circular polarization studying the possible enhancement of decays of arbitrary forbiddenness.

In any case, enhancements will be seen to be of the order of  $(E/E_{crit})^2$  with E being the applied electric field strength. This renders the effects entirely unobservable, given the limited field strength that can for a limited time be obtained under laboratory conditions. In particular, under the conditions envisioned in Ref. 2 the enhancement will be found to be of the order of  $10^{-24}$ . In concluding, we shall discuss the possible reason for the huge discrepancy between Refs. 1, 2 and our results.

# II. BASIC FORMALISM

In order to make this paper self-contained and to introduce our notation we will here outline the basic formalism which has been more extensively presented in a previous paper [3]. We shall exclusively employ the Coulomb gauge and adopt the long wave length approximation for the electromagnetic field so that A = A(t), in accordance with our nonrelativistic treatment. We use units such that A = c = 1, and the electron charge is c = -|c|. To lowest order in the weak interaction gV and to all orders in the electromagnetic interaction the S-matrix element for nuclear beta-decay is

$$S_{fi} = -i \int_{-\frac{\pi}{2}}^{\frac{\pi}{2}} dt \int_{-\frac{\pi}{2}}^{\frac{\pi}{2}} dt \int_{-\frac{\pi}{2}}^{\frac{\pi}{2}} \frac{f(f)^{*}}{(x,t)} \frac{f(f)^{*}}{(x,t)} \frac{f(f)^{*}}{(x,t)}$$

$$* \frac{f}{2} \frac{f(f)^{*}}{(x,t)} \frac{f(f)$$

Here

$$\overline{Y}_{(\nu)}(r,t,q) = e^{-i\left(\overline{E}_{\nu}t - qr\right)}, \quad \overline{E}_{\nu} = 1q$$
(2.2)

is the wave function of the neutrino with momentum q, and

$$\frac{\mathcal{T}_{(e)}(x,t,p)}{-\frac{1}{2m}} = \exp(-i) \left[ Et - pr - e^{2}A^{2}(z) \right] - \frac{1}{2m} \int_{-\infty}^{\infty} dz \left( 2e A(z)p - e^{2}A^{2}(z) \right) \right] \tag{2.3}$$

with  $E = \frac{2}{p}/2m$  is the nonrelativistic Volkov solution describing the electron with momentum p.  $\mathcal{I}^{(i)}$  and  $\mathcal{I}^{(f)}$  are the initial and final nuclear wave functions, which are solutions of a one particle Schrödinger equation

$$i\frac{\partial}{\partial t} \bar{\mathcal{F}}^{(i,f)} = \left[ 2\frac{1}{M_{if}} \left( P - \tilde{e}_{i,f} A \right)^2 + V(r) \right] \bar{\mathcal{F}}^{(i,f)} (2.4)$$

with a self-consistent nuclear potential  $V(\underline{r})$  and reduced charges and masses [1]  $M_{i,f}$  and  $\widetilde{e}_{i,f}$ , respectively. Following Ref. 1, we shall replace the nuclear wave functions by

$$\underline{\underline{Y}}_{0}^{(i,f)} = e^{i \hat{\epsilon}_{i,f}} \underbrace{A(t)}_{\infty}^{r} \underbrace{\underline{J}}_{0}^{(i,f)}$$
(2.5)

with  $\vec{Q_o}$  a solution of the Schrödinger equation with the Hamiltonian

$$\mathcal{H}_{0} = \frac{\mathbb{R}^{2}}{2 M_{i,f}} + V(\gamma) . \qquad (2.6)$$

The wave function (2.5) would be exact if  $\oint_0$  were derived from the Hamiltonian  $H_0 - \operatorname{erE}(t)$  with  $E = -\partial A/\partial t$ , rather than from Eq. (2.6). It is important to note that the wavefunction (2.5), as it stands, does not include, contrary to appearance, any interaction with the external field, but is rather the correct noninteracting wave function in the Coulomb gauge. It is here that our interpretation deviates from Ref. 1, where it is assumed that the  $\bigvee_0^{(v,f)}$  represent a fair approximation to the interacting nuclear wave functions. We have previously discussed this point at length [3]. Here we shall be content with pointing out that the operator  $p = -i \bigvee_0^v \ln Eq$ . (2.6) represents the canonical momentum p = Nv + eA, which does not, since we use the Coulomb gauge with  $A \neq 0$ , agree with the mechanical momentum. Hence an eigenfunction of  $H_0$  does not appropriately describe the state in the absence of the field. This is evident from the expectation value of the velocity in an eigenstate of  $H_0$ .

which does depend on the field, whereas the expectation value in a state (2.5),

does not, as it should be for a noninteracting state. Hence one has to be aware that by using Eq. (2.5) one entirely ignores the interaction between the nucleus and the external field.\*

The total decay rate per unit time is obtained from

$$\overline{I} = \int \frac{d^3p}{(2\pi)^3} \frac{d^3q}{(2\pi)^3} \frac{f_{\text{DM}}}{T_{-\infty}} \frac{1}{\sqrt{15}} \int_{f_2}^{2\pi} \frac{1}{\sqrt{15}} \frac$$

The standard procedure is now to first evaluate the differential decay rate  $|S_f|^2$  and then to do the integration over phase space. This would require to specify the external field A(t) from the outset so that the integration over time in  $S_f$  can be carried out. The final integration over phase space is formidable [1,4,5] and has not been done yet in Refs. 4 and 5. Since we are not interested in the differential decay rate, we shall here reverse the older of integrations: we shall first integrate over phase space expressing the total decay rate in terms of the electron's and neutrino's Green functions, and do the integrations over time afterwards. The integration over f in f need not concern us here; it will be absorbed into a nuclear matrix element which is left untouched. Hence we write

<sup>\*</sup> In view of the fact that the frequency and field strength of the external field are very small on a nuclear scale one would expect this to be at least a fair approximation. Unfortunately, no exactly solvable model seems to be available in order to test this assumption. The only exception is the harmonic oscillator which can be solved exactly in the presence of a periodic external field. This solution corroborates the above assumption. However, the harmonic oscillator is not a very realistic example, given the equidistance of its levels and the lack of a continuum. So the question of whether the field-nucleus interaction might produce noticeable effects, remains open to some extent, in particular, if the nucleus has a very low lying excited state or if he nuclear energy release in the beta decay is very small. In any event, to improve on the approximation (2.5) will be very hard.

$$\Gamma = \int \alpha^3 r \, \dot{\alpha}^3 r' \, d^3 r' \, d^$$

with

$$y(x,x') = \frac{1}{\sqrt{\pi}} \int_{0}^{\pi} \int_{$$

In the preceding equation we introduced  $E_0 = E_1 - E_{\uparrow} - m$  as the nuclear energy release minus the electron mass and have taken the charge difference [1]  $e_1 - e_{\uparrow}$  to be e, as required by charge conservation. The Green functions are

$$G_{(\nu)}(\tau't', \tau t') = \int_{\alpha}^{2} \int_{\gamma}^{2} \frac{1}{(\nu)(\tau't'; 2)} \frac{1}{2} \frac{1}{(\nu)(\tau't'; 2)}$$

$$= \frac{4\pi}{x} \int_{0}^{\infty} cl E_{\nu} E_{\nu} e^{-iE_{\nu}(\tau-iE)} \int_{\gamma}^{2} \frac{1}{(\tau+x-iE)^{2}} \frac{1}{2} = G_{(\nu)}(\tau,x)_{(2.10)}$$

with

$$\tau = t' - t \qquad , \qquad x = \int_{\infty}^{\infty} \tau' - \tau' \qquad , \qquad (2.11)$$

and

$$G_{(e)}(\tau't',\tau t) = \int_{0}^{0} \int_{-\infty}^{\infty} \underbrace{T_{(e)}(\tau,t;R)} \underbrace{T_{(e)}(\tau't';R)}$$

$$= e \underbrace{\tau'-\tau}_{t'-t} \int_{-\infty}^{\infty} A(\xi) d\xi, \quad -i \mathcal{M}(t',t)$$

$$= e \underbrace{G_{(e)}(\tau,t)}_{(e)}(\tau,t)$$

$$= e \underbrace{G_{(e)}(\tau,t)}_{(e)}(\tau,t)$$

where

$$\mathcal{M}(t,t) = \frac{e^{2}}{2m} \left\{ \int_{t}^{t} d\xi, A^{2}(\xi) - \frac{1}{2} \left( \int_{t}^{t} d\xi, A^{2}(\xi) \right)^{2} \right\}.$$
(2.13)

In Eq. (2.12) we introduced the free electron Green function

$$G_{(e)}(\tau,x) = \frac{4\pi m}{x} \int_{0}^{\infty} c(E e^{-iE(T-iE)} \sin x \sqrt{2mE})$$

$$= \left(\frac{2\pi \pi}{i(T-iE)}\right)^{\frac{3}{2}} e^{-im\frac{x^{2}}{2\tau}}$$
(2.14)

The Green functions allow for explicit representations as exhibited in the last lines of Eqs. (2.10) and (2.14); however, the preceding integral representations with respect to energy will turn out to be more convenient.

Noting that

$$e\left(A(t)x - A(t')x'\right) - \frac{e(x'-x)}{t'-t} \int_{t}^{t'} d\xi, A(\xi)$$

$$= e \int_{t}^{t} d\xi, R(\xi) E(\xi) = R(t',t)$$

$$= \frac{1}{2} \left(\frac{t}{t}\right) \left(\frac{t}{t}\right)$$

$$= \frac{1}{2} \left(\frac{t}{t}\right)$$

with

$$R(\xi) = \chi' - \frac{\chi' - \chi}{t' - t} (t' - \xi)$$
(2.16)

we can now rewrite Eq. (2.9) as

$$Y(x, x') = \frac{1}{(2\pi)^6} \lim_{T \to \infty} \frac{1}{T} \int_{-T/2}^{T/2} i R(t/t)$$

$$\times e^{-i M(t/t)} i E_0 T G(y, (T/x)) G(y/t/x). \qquad (2.17)$$

This representation has some pleasant features. The entire field dependence is contained in the functions  $\mathcal{R}$  (t',t) and  $\mathcal{M}$ (t',t) so that by dropping them we recover the free decay rate (for an arbitrary degree of forbiddenness). The quantity  $\mathcal{R}$  is more than just a convenient abbreviation. It is the only term which depends both on the field and on r and r'. Hence it is the prime candidate for a possible field induced removal of forbiddenness. However, Eq. (2.15) shows remarkable cancellations: all the individual terms on the left hand side are of the order of  $e|A|R_o$  with  $R_o$  the nuclear radius. For the experimental conditions envisioned in the introduction this can readily be of order unity or even larger. In contrast, the right hand side involves the

electric field E rather than the vector potential, and we have

$$\mathcal{R}(t',t) \sim e |A| R_0 \omega r$$
 (2.18)

If we assume that the integrals over time receive significant contributions only for  $|\mathcal{T}| = |\mathbf{t'} - \mathbf{t}| \lesssim E_0^{-1}$  as suggested by the uncertainty relation, we have in Eq.  $(2.18) \, \omega \, \mathcal{T} \approx \omega / E_0 \ll 1$  so that the impact of the function  $\mathcal{R}(\mathbf{r'},\mathbf{t})$  should be marginal. Even if we do not invoke the uncertainty relation, we notice from Eqs. (2.10) and (2.14) that the integrand of Eq. (2.17) is proportional to  $\mathcal{T}^{-9/2}$ . Notably, this rapid drop-off is independent of the external field. This fact makes it very unlikely that the integral (2.17) will receive any significant contributions from larger values of  $\mathcal{T}$ . With regard to the second field-dependent function in Eq. (2.17) we note its power series expansion

$$\mathcal{M}(t/t) = \frac{\tau^{2}}{24m} \left( e E\left(\frac{t+t}{2}\right) \right)^{2} + \dots$$
 (2.19)

This starts with the cubic power of  $\tau$ , so that it is not likely to play any significant role, either.

The preceding arguments suggest already that any field induced effects should be fairly small. Yet, hand waving estimates of the double integral (2.17) might seem to be unreliable. Because of that, and in order to get explicit numbers, we shall turn to an explicit evaluation in the next Section. III. EXPLICIT CALCULATTION OF THE TOTAL DECAY RATE FOR CIRCULAR POLARIZATION We shall now evaluate Eq. (2.17), explicitly for a monochromatic circularly polarized field

$$A(t) = \alpha \left( e_{x} \cos \omega t + e_{y} \sin \omega t \right).$$
(3.1)

With slightly more algebra, linear polarization could be treated along the same lines. The field dependent functions (2.13) and (2.15) are now

$$\mathcal{R}(t,t) = ea \left\{ \left( \frac{\sin(\omega t/2)}{\omega t/2} - \cos(\omega t/2) \right) \right.$$

$$\times \left( (x'-x) \cos \omega t' + (y'-y) \sin \omega t' \right)$$

$$+ \sin(\omega t/2) \left( (x'+x) \sin \omega t' - (y'+y) \cos \omega t' \right) \left. \right\}$$

$$(3.2)$$

and

$$\mathcal{M}(t,t) = \frac{(\epsilon a)^2}{2m} \tau \left( 1 - \left( \frac{\sin k \tau/2}{k \tau/2} \right)^2 \right) \equiv \mathcal{M}(\tau). \quad (3.3)$$

Note that  $\mathcal{M}(t',t)$  does only depend on  $\mathcal{T}$ , and the integrand of Eq. (2.17) only depends on  $\mathcal{T}'=(t+t')/2$  via the quantity  $\mathcal{K}(t',t)$ . The integral over  $\mathcal{T}'$  can now be done:

$$\lim_{T\to\infty}\frac{f_{2}}{f}\int_{-\overline{I}_{2}}^{\overline{I}_{2}}e^{i\mathcal{R}(t',t)}=\overline{J_{0}\left(ea\sqrt{\overline{z}(\overline{t})}\right)}$$

$$(3.4)$$

with

$$\frac{2(\tau)}{(2\tau)} = \left(\frac{2\tau}{2\tau} - \frac{2\tau}{2\tau}\right)^{2} \left[ \frac{\sin((\omega\tau/2))}{(\omega\tau/2)} - \cos((\omega\tau/2)) \right]^{2} \\
+ \left(\frac{2\tau}{2\tau} + \frac{2\tau}{2\tau}\right)^{2} \left[ \frac{\sin((\omega\tau/2))}{(\omega\tau/2)} - \cos((\omega\tau/2)) \right]^{(3.5)} \\
+ \left(\frac{2\tau}{2\tau} + \frac{2\tau}{2\tau}\right)^{2} \sin^{2}((\omega\tau/2)),$$

where  $r_{\tau} = (x,y)$ ,  $r'_{\tau} = (x',y')$ . Eq. (2.17) can now be rewritten as

$$f(x,x') = e^{-i\partial x(-i\partial/\partial \bar{\epsilon}_0)} \sqrt{e^{(2(-i\partial/\partial \bar{\epsilon}_0))}}$$

$$\times f_0(x,x')$$

$$\times f_0(x,x')$$
(3.6)

where f is the field independent part of Eq. (2.17), i.e., the right hand side with  $\mathcal{R}$  (t',t) =  $\mathcal{M}$ (t',t) = 0. In Eq. (3.6) we exploited the particular Equation dependence of Eq. (2.17) in order to replace  $\mathcal{L}$  by  $-i\mathcal{L}$  in the field dependent terms. Hence we could pull the latter terms out of the integral over  $\mathcal{L}$ . No assumption as to the smallness of  $\mathcal{L}$  is involved. Hence, barring a couple of differentizations, the problem is reduced to the field free one.

The quantity  $\int_{c}^{c} (r,r')$  is now most easily evaluated by inserting the integral representations (2.10) and (2.16) and expanding everything with respect to x = |r - r'|. The result is

$$V_{0}(x, x') = V_{0}(x) = \int_{\mathbb{R}^{n}} \int_{\mathbb{R}^{n}} \int_{-\infty}^{\infty} G(y, (\xi x)) G(y, (\xi x)) G(y, (\xi x)) G(y, (\xi x))$$

$$= \frac{8\sqrt{2}}{\pi^{3}} m^{3} \int_{\mathbb{R}^{n}} \int_{\mathbb{R}^{n}}^{\infty} (-E_{0} m x) \int_{\mathbb{R}^{n}}^{\infty} \frac{(l+1)(4E_{0}/m)!}{(k-l)!(2k+2l+7)!!} \frac{1}{(3.7)}$$

One of the two sums can be carried out in a multitude of ways, but the explicit form (3.7) is the most useful one.

We now have to insert Eq. (3.7) into Eq. (3.6) and to carry out the required differentiations. Needless to say, Eq. (3.6) would be entirely useless if the differential operators could not be expanded into power series with respect to  $\partial/\partial E_0$ . We shall first write down and then discuss the expansions of exp (-i) $\pi$ ) and  $\sqrt{\partial}$  (ea  $\sqrt{Z}$ ); with the abbreviations

$$\alpha = \frac{\omega \tau}{2} = -\frac{i\omega}{2\partial E_0}, \qquad l = \frac{(e\alpha)^2}{m\omega}, \qquad (3.8)$$

they are

$$e^{-i\frac{3\pi(2)}{3}} = 1 - i\frac{1}{3}\left(1 - \frac{2\alpha^{2}}{15} + \frac{\alpha^{4}}{105} + \dots\right)$$

$$-\frac{1^{2}\alpha^{6}}{18}\left(1 - \frac{4\alpha^{2}}{15} \dots\right) + O(1^{3}\alpha^{9})$$
(3.9)

and

$$J_{0}\left(ea\sqrt{z}\right) = 1 - \frac{(ea)^{2}}{4} \left[ x^{2}(x_{T} + x_{T}^{2})^{2}(1 - x_{T}^{2} + \dots) + \frac{1}{9}(x_{T}^{4} + \dots)(x_{T}^{2} - x_{T}^{2})^{2} + \frac{4}{3}x^{3}(x_{Y}^{2} - x_{Y}^{2})(1 - \frac{4x^{2}}{15} + \dots) + \frac{(ea)^{4}}{64}(x_{T}^{4} + x_{T}^{2})^{4} + 0\left(eax(x_{T}^{4} + x_{T}^{2})\right)^{6}.$$
(3.10)

In applying the differential operators (3.9) and (3.10) to Eq. (3.7), we first note that each  $\propto$  will effectively be replaced by a very small number,

$$\alpha E_0^{\nu} = -\frac{i\nu}{2} \frac{\omega}{E_0} E_0^{\nu}$$

since the ratio  $W/E_0$  is very small in any case; for  $E_0 \sim m$  we have  $W/E_0 \sim 10^{-6}$  for a laser field and  $W/E_0 \sim 10^{-14}$  for the radio frequency fields with  $J \sim 100m$ 

which are considered in Ref. 2. The effective expansion parameter in Eq. (3.9) is then

$$\lambda x^{3} \sim \frac{(ea)^{2}}{m\omega} \left(\frac{b}{\overline{\epsilon}_{0}}\right)^{3} = \left(\frac{m}{\overline{\epsilon}_{0}}\right)^{3} \left(\frac{\overline{\epsilon}}{\overline{\epsilon}_{crit}}\right)^{2}$$
(3.11)

and

$$(eaR_0x)^2 \sim (eaR_0\frac{L_0}{E_0})^2 = (\frac{m^2R_0}{E_0})^2 (\frac{E}{E_{crit}})^2$$
 (3.12)

in Eq. (3.10) with  $R_0 \sim 1.1 \times 10^{-13}$  cm x A the nuclear radius. Both quantities are very small for experimentally available fields so that the expansions (3.9) and (3.10) are very well justified. With respect to Eq. (3.7) we note that the main expansion parameter

$$E_0 m x^2 \lesssim E_0 m (2R_0)^2 \simeq 3.6 \times 10^{-5} A^{\frac{2}{3}} \frac{E_0}{m}$$
 (3.13)

is again small. Hence, it will be sufficient in most applications to keep only the lowest power of x which is necessary in view of the forbiddenness of the decay. The sum over  $\ell$  is then restricted to a few terms.

Let us first consider the field induced enhancement of allowed beta decay. Since the expansion parameter (3.12) is in general smaller than (3.11) we can ignore  $J_{c}(ea\sqrt{z})$ . Keeping only the lowest order terms in Eq.(3.9) and using Eq.(3.7) (with x = 0), (3.6) and (2.8) we arrive at

$$\Gamma = \left| \left\langle \overline{\Phi}^{(f)} \right| g V \left| \overline{\Phi}^{(i)} \right\rangle \right|^{2} \frac{\delta V \overline{\lambda} m^{\frac{3}{2}} \overline{E}_{0}^{\frac{3}{2}}}{105 \pi^{3}}$$

$$\times \left\{ 1 + \frac{35}{8} \left( \frac{m}{2\overline{E}_{0}} \right)^{3} \left( \frac{\overline{E}}{\overline{E}_{crit}} \right)^{2} \left( 1 - \frac{1}{30} \left( \frac{l_{V}}{2\overline{E}_{0}} \right)^{2} \right) + \frac{35}{128} \left( \frac{m}{2\overline{E}_{0}} \right)^{6} \left( \frac{\overline{E}}{\overline{E}_{crit}} \right)^{4} \right\}. \tag{3.14}$$

Apparently, the field induced enhancement is unobservably small\*. In Eq. (3.14) we included the next-to-leading terms in order to be able to make the comparison with the nonrelativistic limit of a fully relativistic calculation [13,14] of laser enhancement of allowed beta-decay. Equation (3.14) agrees with Eq. (113) of Ref. 14. Actually, the next-to-leading terms as exhibited in Eq. (3.14) are incomplete, because additional terms resulting from  $J_{g}$  (ea  $\sqrt{z}$ ) (Eq. (3.10)) are of at least the same order of magnitude. Since the leading term is extremely small already, we did not care to write them down.

We will now turn to a first order forbidden decay. In the absence of the field the leading contribution to the decay rate comes from the term with k=1 in Eq. (3.7). Recalling that  $x^2 = (r - r')^2$  and that we need one power of r in each nuclear matrix element to render if nonzero, we obtain

$$\Gamma_{i} = \left| \left\langle \vec{\Phi}_{0}^{(i)} / g V_{x} / \vec{\Phi}_{0}^{(i)} \right\rangle \right|^{2} \\
\times \frac{16 \sqrt{2} m^{5/2} \vec{E}_{0}^{3/2}}{945 \pi^{3}} \left( 1 + \frac{d \vec{E}_{0}}{1/m} \right) \tag{3.15}$$

<sup>\*</sup> Previous conclusions to the contrary [10,11] were flawed and due to an inadequate approximation in evaluating sums over Bessel functions. The present authors have corrected this mistake in the general form of a no-go theorem [12].

Considering now the external field we know already from the previous example that the enhancement with is formally due to the operator  $\mathcal{N}_c$ , is extremely small. Hence, we shall rather consider the operator  $J_0(ea\sqrt{z})$  which, via its dependence on r and r', generates the kind of field induced modifications of the selection rules which are proposed in Refs. 1 and 2. Hence in the presence of the field we do obtain a non zero contribution to the decay rate from the term with k = 0 in Eq. (3.7), which we shall call

$$\Gamma_{0,\alpha} = \int \alpha^{3} r \, d^{3} r' \left\{ \int_{0}^{f} f'(x) \int V \int_{0}^{f} f'(x) \right\}$$

$$\times \left\{ \int_{0}^{f(i)} f'(x) \int V \int_{0}^{f} f'(x) \int V \int_{0}^{f} (e\alpha \sqrt{2}) \left[ \int f'(x, x') \right]_{x=0} \right\}$$
(3.16)

Keeping just the leading term in the expansion (3.10) we get\*

$$\vec{l}_{0,\alpha} = |\langle \vec{\phi}_{c}^{(f)} | \hat{g} | \nabla_{x_{f}} | \vec{\phi}_{0}^{(i)} \rangle|^{2} \sqrt{2} m^{\frac{3}{2}} \vec{\epsilon}_{0}^{\frac{3}{2}} (\epsilon_{0} \omega)^{2} \frac{1}{2} \vec{\epsilon}_{0}^{\frac{3}{2}} (\epsilon_{0}$$

If we assume that  $\langle r_T \rangle / \langle r \rangle = 2/3$ , the enhancement is

$$\frac{\Gamma_{0,2}}{\Gamma_{1}} = \frac{105}{32} \frac{(ea \omega)^{2}}{m E^{2}} \left(1 + \frac{\delta \overline{c}_{0}}{11m}\right)^{-1}$$

$$= \frac{105}{32} \left(\frac{m}{\overline{\epsilon}_{0}}\right)^{3} \left(\frac{\overline{E}}{\overline{\epsilon}_{crit}}\right)^{2} \left(1 + \frac{\delta \overline{\epsilon}_{0}}{11m}\right)^{-1}.$$
(3.18)

<sup>\*</sup> The last term in the square bracket in Eq. (3.10) seems at first glance to yield an imaginary contribution to the decay rate. Actually it cancels when integrated over with the nuclear wave functions.

Hence we arrive at much the same conclusion as in the case of allowed beta-decay: the enhancement which is due to field induced modifications of the selection rules is by far too small to be observable.

In order to obtain the total enhancement of a first forbidden decay we still have to add the modification of  $\widehat{I}_{r}$ , due to the operator  $\partial I_{r}$ , viz.

$$\Gamma_{,a} = |\langle \bar{\psi}_{o}^{(f)} | g V_{x} | \bar{\psi}_{o}^{(i)} \rangle|^{2} \sqrt{2} m^{\frac{J_{2}}{J_{2}}} \bar{\psi}_{o}^{\frac{J_{2}}{J_{2}}} (ea(u)^{2})^{2} \times (/ + \frac{4\bar{t}_{o}}{5m}), \qquad (3.19)$$

so that the total enhancement is

$$\frac{\Gamma_{0,a} + \Gamma_{1,a}}{\Gamma_{1}} = \frac{231}{64} \frac{15m + 2E_{0}}{11m + 2E_{0}} \left(\frac{m}{E_{0}}\right)^{3} \left(\frac{E}{E_{crit}}\right)^{2}. \quad (3.20)$$

To give a specific example, let us look at the first order forbidden decay of  $^{90}$ Sr which was considered in Ref. 2. In that paper an enhancement of the half life from the usual 29 years to about 10 years is predicted to occur in a radio frequency field environment with E  $\sim 8 \times 10^{3}$  Volt/cm. Corrected estimates have not been given yet, but it is argued [4,5] that enhancements of first order forbidden decays should stay more or less like they were. In contrast, Eq. (3.20) yields for this case a relative change in the decay rate of about  $\frac{-24}{2} \times 10^{3}$ .

The preceding calculation was for circular polarization. With slightly more labor, it can be done for linear polarization as well, with qualitatively the same results. Moreover, it can be shown that for an arbitrary plane wave field (arbitrary polarization and pulse shape) the total decay rate is entirely independent of the field as long as the quasiclassical limit applies [15].

# IV. CONCLUSIONS

We have shown that the effect of an intense plane wave field on forbidden beta-decay is much the same as it is on allowed beta-decay. There are two types of field-induced contributions to the total decay rate: one that does, via its dependence on the nuclear coordinate, change the selection rules of the decay and one that does not. Both terms lead to an enhancement of the decay, but in either case the enhancement is proportional to the squared ratio  $(E/E_{cnit})^2$  of the applied over the critical field  $E_{cnit} = 1.3 \times 10^{-5}$  Volt/cm. For an intense focused laser field we have  $E/E_{cnit} \lesssim 10^{-5}$  at best, for a radio frequency field the ratio is very much smaller.

Our results for the enhancement differ from previous recent estimates [1,2,4,5] by twenty-four orders of magnitude. The reason for this large discrepancy seems to be the following: the original explicit estimates for various specific examples [2] were later shown to be faulty [3] and withdrawn [4,5]. However, in Refs. 4 and 5 the conclusion is maintained that there should be an enhancement of the order of unity without explicit numbers being given. This conclusion is drawn from the observation that there is a significant effect of the external field on the differential decay rate. This, however, does not necessarily reflect any substantial changes of the total decay rate. We have shown previously [12] that the total decay rate of a neutral particle decaying into various charged particles in the presence of an intense plane wave field, is in the semi-classical (WKB) limit entirely independent of the field. This is notwithstanding the fact that the energy spectrum of the decay products is completely distorted by the field as compared to the field-free one ( for examples in the case of allowed beta-decay see Ref. 13). The latter effect is due to the essentially classical interaction between the field and the charged decay products once the decay has already taken place. The decay itself is not affected until the field becomes comparable with the (genuinely quantum

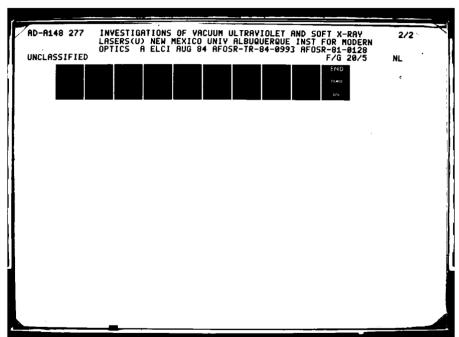
mechanical) critical field  $\mathbf{E}_{\mathrm{Crit}}$ . Reference 12 does not strictly apply to the present situation because the decaying nucleus is not neutral and there is no allowance for bound particles. It is, however, easily extended to the present situation [15]. The bottom line is the same: enormous effects on the differential decay rate do not correspond to any significant enhancement of the total rate. The calculation initiated in Refs. 1, 2, 4, and 5 should, if finished, lead to the same conclusion.

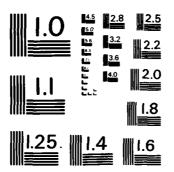
We should add a critical remark regarding our calculational approach exhibited in Section 4. Equation (3.6) which displays the decay rate as the result of applying a differential operator to the free decay rate is exact as it stands. However, the following ev luation proceeds via perturbation theory with respect to the external field. This is at a stage where the quantitative legitimacy of perturbation theory is obvious; yet, the exact result is likely to contain terms which are quantitatively minute, but nonanalytic with respect to the coupling to the external field. This is known to be the case for allowed beta-decay [13,14]. These terms cannot be obtained by our procedure. It should be emphasized that we do not apply perturbation theory to the differential decay rate; the latter containing multiphoton interactions of extremely high order (up to 10 between the field and the electron is clearly inaccessible by perturbation theory of any reasonable finite order. However, the total decay rate is a different matter: for fields below the critical field perturbation theory yields a safely convergent asymptotic series.

We mention, finally, that the situation is entirely different, if there is a significant interaction between the nucleus and the field. This happens, for instance, if an intense x-ray laser is nearly resonant with an excited nuclear level. In this case substantial enhancements of the total decay rate are found [16].

# REFERENCES

- 1. H. R. Reiss, Phys. Rev. C27 (1983) 1199.
- 2. H. R. Reiss, Phys. Rev. C27 (1983) 1229.
- 3. W. Becker, R. R. Schlicher and N. O. Scully, to be published in Physic Fev. C.
- 4. H. R. Reiss, Phys. Rev. C23 (1983) 1402.
- 5. H. R. Reiss, to be published in Phys. Rev. C.
- 6. E. Fermi, Zs. Phys. 88 (1934) 161.
- 7. G. T. Emery, Ann. Rev. Nucl. Sci. 22 (1972) 165.
- 8. G. C. Baldwin, S. A. Wender, Phys. Rev. Lett. 48 (1982) 1461.
- 9. J. Schwinger, W. Y. Tsai, and T. Erber, Ann. Phys. (N.Y.) 96 (1976) 303.
- 10. I. G. Baranov, Izv. Vyssh. Uchebn. Zaved. Fiz, No. 4 (1974) 115.
- 11. W. Becker, W. H. Louisell, J. D. McCullen, and M. O. Scully, Phys. Rev. Lett. 47A (1981) 1262.
- 12. W. Becker, G. T. Moore, R. R. Schlicher, and M. O. Scully, Phys. Lett. <u>94A</u> (1983) 131.
- 13. I. M. Ternov, V. N. Rodicnov, A. E. Lobanov, and O. F. Dorofeyev, Pis'ma Zh. Eksp. Teor. Fiz. 37 (1983) 288 [JETP Lett. 37 (1983) 342]; I. M. Ternov, V. I. Rodinov, and O. F. Dorofeyev, Zh. Eksp. Teor. Fiz. <u>84</u> (1983) 1225.
- 14. A. I. Nikishov, V. I. Ritus, Zh. Eksp. Teor. Fiz. 85 (1983) 24.
- 15. W. Becker, R. R. Schlicher, and M. O. Scully, to be published.
- 16. W. Becker, R. R. Schlicher, M. O. Scully, M. S. Zubairy, and M. Goldhaber, Phys. Lett 1318 (1983) 16.





MICROCOPY RESOLUTION TEST CHART NATIONAL BUREAU OF STANDARDS -1963 - A

# Comment on enhancement of forbidden nuclear beta decay by high-intensity radio-frequency fields

W. Becker, R. R. Schlicher, and M. O. Scully
Max-Planck Institut für Quantenoptik, D-8046 Garching bei München, West Germany
and Institute for Modern Optics, Department of Physics and Astronomy,
University of New Mexico, Albuquerque, New Mexico 87131
(Received 23 May 1983)

A recent claim that forbidden nuclear beta decay can, by the application of a high-intensity radio-frequency field, be enhanced by many orders of magnitude is contested. The effect is shown to be nonexistent, at least within the theoretical model which has been adopted thus far.

### I. INTRODUCTION

In two interesting recent publications<sup>1,2</sup> Reiss claims that forbidden nuclear beta decay can, in the presence of an intense, but readily achievable radio frequency field, be enhanced by many orders of magnitude, the more so the higher forbidden the decay is. Of course, this would be a fascinating effect, both from a theoretical and a practical point of view. Unfortunately, we find, at least in the framework of the approximations and idealizations underlying this work, his conclusions to be incorrect and the effect nonexistent.

The approach of Ref. 1 to nuclear beta decay in the presence of a strong external electromagnetic field treats the weak coupling to first order of perturbation theory, but is supposed to take into account the coupling with the external field to all orders, at least approximately. Hence in Reiss's model the state of the emitted electron is described by the Volkov wave function,3 whereas the nuclear states are to be well approximated by the so-called momentum translation approximation (MTA) wave function. We shall show in Sec. II by explicit calculation that within this particular model the total beta-decay rate is essentially independent of the external electromagnetic field. Our argument turns out to be independent of whether or not we employ a relativistic description of the decay. Hence, for the sake of simplicity, we shall first turn to a completely nonrelativistic description within a long wavelength approximation for the applied field. Once we have made our point, the generalization to the fully relativistic problem is conceptually straightforward and will be presented in Sec. III. Our conclusion that the lifetime of a nucleus cannot be influenced by an external field in the framework of the model adopted by Reiss leads us to suspect that Ref. 1 includes a calculational error. In Sec. IV we point out such an error. In order to understand

why Reiss's model cannot yield an enhancement of the nuclear decay rate we critically review the derivation and justification of the MTA wave function in Sec. V. By comparing the "r·E" vs "p·A" form of the interaction Hamiltonian we obtain an indication of why the MTA method is an unjustified approximation. We then point out with the help of gauge arguments<sup>5</sup> that the MTA wave function is nothing but the correct unperturbed state in the Coulomb gauge.6 This statement can be corroborated by writing the S matrix for the beta decay in different gauges. In this way we see that in Reiss's theory only the interaction of the field with the electron is incorporated exactly, whereas the interaction with the nucleus is completely neglected. This fact is in strict contrast to the statements and intentions of Ref. I and leads to a completely different interpretation of the model. If the interaction of the nucleus with the external field is not incorporated into the model, a modification of the total decay rate can only originate from the coupling of the electron to the field. It was recently shown that decays of neutral particles are unaffected by the application of optical and, even more so, radio frequency fields, to an excellent approximation. Consequently, since Reiss's approach to the problem does not contain any genuine interaction of the nuclei with the field, it cannot yield any impact on the nuclear lifetime. In Sec. VI we summarize our various criticisms of Ref. 1.

# II. FIELD INDEPENDENCE OF THE TOTAL TRANSITION PROBABILITY

The starting point of the formalism of Refs. 1 and 2 is the S matrix for nuclear beta decay. If we denote the weak interaction, which causes the transition, by (gV), the nonrelativistic limit to the S-matrix element to first order in the weak coupling reads

$$S_{p} = -i \int d^{3}r \int dt \, \Psi_{\text{MTA}}^{(f)*}(\vec{r},t) \Psi_{(e)}^{*}(\vec{r},t;\vec{p}) \Psi_{(r)}^{*}(\vec{r},t;\vec{q})(gV) \Psi_{\text{MTA}}^{(f)}(\vec{r},t) . \qquad (2.1)$$

The various terms in Eq. (2.1) are discussed below.  $\Psi_{(\nu)}$  denotes the neutrino wave function, which is a plane wave with momentum  $\vec{q}$ . Following the procedure outlined in Ref. 1, we take the wave functions of the charged particles, i.e., the electron and the nucleus in the initial and final state, to be in the Coulomb gauge. The electron wave func-

tion is then a solution of the Schrödinger equation

$$i\frac{\partial}{\partial t}\Psi_{(e)}(\vec{r},t) = \frac{1}{2m}[\vec{p} - e\vec{A}(t)]^2\Psi_{(e)}(\vec{r},t) . \qquad (2.2)$$

We use here natural units  $\hbar - c - 1$ . Unlike the convention in Ref. 1 where e - |e|, we denote the electric charge of a

(2.9)

particle by e, so that, for instance, the electron charge is e = -|e|. The exact solution of Eq. (2.2) is the nonrelativistic Volkov wave function with momentum  $\vec{p}$ ,

$$\Psi_{(\mathbf{e})}(\vec{r},t;\vec{p}) = \exp\left[-i\left[Et - \vec{p}\vec{r} - \frac{1}{2m}\int^{t} [2e\vec{A}(\tau)\vec{p} - e^{2}\vec{A}^{2}(\tau)]d\tau\right]\right], \qquad (2.3)$$

where  $E = p^2/2m$ , so that our nonrelativistic theory is only consistent for low energy emitted electrons.

The underlying nuclear model of Ref. 1 is the shell model with an inert  $0^+$  core, which is affected neither by the beta decay nor by the electromagnetic field, and one or more valence nucleons in an angular momentum coupled state. The nuclear wave functions  $\Psi^{(r)}$  and  $\Psi^{(f)}$  are then derived from a one-particle Hamiltonian. The initial and final nuclear states are approximated in Ref. 1 by the so-called momentum translation approximation (MTA) wave function. This wave function is given by

$$\Psi_{\text{MTA}}(\vec{r},t) = \exp[i\tilde{e}_{N}\vec{A}(t)\vec{r}]\Phi_{0}(\vec{r},t) , \qquad (2.4)$$

where  $\tilde{e}_N$  is the reduced nuclear charge [Eq. (6) of Ref. 1] and  $\Phi_0$  denotes the nuclear wave function in the absence of the external electromagnetic field, i.e.,

$$i\frac{\partial}{\partial t}\Phi_0(\vec{r},t) = H_0\Phi_0(\vec{r},t) , \qquad (2.5)$$

with

$$H_0 = \frac{1}{2m_*} \vec{p}^2 + V(\vec{r}) . {(2.6)}$$

 $V(\vec{r})$  denotes the nuclear binding potential, and  $m_r$  is the reduced nuclear mass [Eq. (5) of Ref. 1]. Since we consider the nucleus initially and finally to be in an eigenstate  $\phi_n(\vec{r})$  (n = i, f) of  $H_0$  with energy  $E_n$ , we shall later use  $\Phi_0$  in the form

$$\Phi_0^{(n)}(\vec{r},t) = e^{-iE_n t} \phi_n(\vec{r}) \quad . \tag{2.7}$$

For the time being we shall adopt the MTA wave function in the same way, as it is used in Ref. 1, and postpone a detailed discussion on this subject to Sec. V.

The total transition probability per unit time is calculated from the S-matrix element (2.1) by

$$\Gamma = \int \frac{d^3q}{(2\pi)^3} \int \frac{d^3p}{(2\pi)^3} \lim_{T \to \infty} \frac{1}{T} |S_{fi}|^2 . \tag{2.8}$$

If we insert the S-matrix element (2.1) into Eq. (2.8) using the wave functions (2.3), (2.4), (2.7), and plane waves for the neutrino, the total rate  $\Gamma$  takes the form

$$\Gamma = \frac{1}{(2\pi)^6} \lim_{\vec{r} \to \infty} \frac{1}{T} \int d^3r \, d^3r' \int_{-T/2}^{T/2} dt \, dt' \exp[iE_0(t'-t)] \exp\{-ie[\vec{A}(t')\vec{r}' - \vec{A}(t)\vec{r}']\}$$

$$\times G_{(\nu)}(\vec{r}',t';\vec{r},t) G_{(e)}(\vec{r}',t';\vec{r},t)$$

 $\times [\phi_{\ell}^*(\vec{r})(gV)\phi_{\ell}(\vec{r})][\phi_{\ell}^*(\vec{r}')(gV)^*\phi_{\ell}(\vec{r}')] .$ 

Here  $E_0 = E_i - E_f$  denotes the nuclear energy released in the beta decay. We furthermore used the relation  $\tilde{e}_i - \tilde{e}_f = e$  [see Ref. 1, Eq. (49), and our convention for the sign of e]. In fact, this relation is only approximate, since the minor impact of the external field on the nuclear core is neglected. The sum over the neutrino states is expressed in terms of a plane wave Green's function with a dispersion  $E_e = |\vec{q}|$ .

$$G_{(p)}(\vec{r}',t';\vec{r},t) = \int d^3q \, \Psi_{(p)}^*(\vec{r},t;\vec{q}) \Psi_{(p)}(\vec{r}',t';\vec{q}) = \int d^3q \, \exp[-i|\vec{q}|(t'-t)] \exp[i\vec{q}(\vec{r}'-\vec{r})] \,. \tag{2.10}$$

All the electronic contributions to the total rate  $\Gamma$  are contained in the nonrelativistic Volkov Green's function

$$G_{(e)}(\vec{r}',t';\vec{r},t) = \int d^3p \Psi_{(e)}^{\bullet}(\vec{r},t;\vec{p}) \Psi_{(e)}(\vec{r}',t';\vec{p})$$

$$= \exp \left\{ -i \frac{e^2}{2m} \int_t^{t'} \vec{A}^2(\tau) d\tau \right\} \int d^3p \exp \left\{ -i \left[ \frac{t'-t}{2m} \vec{p}^2 - \left[ \vec{r}' - \vec{r} + \frac{e}{m} \int_t^{t'} \vec{A}(\tau) d\tau \right] \vec{p} \right] \right\}. \tag{2.11}$$

The integral (2.11) can be reduced to a Gaussian integral and we find

$$G_{(e)}(\vec{r}',t';\vec{r},t) = \left(\frac{2m\pi}{i(t'-t)}\right)^{3/2} \exp\left[i\frac{m}{2} \frac{(\vec{r}'-\vec{r})^2}{t'-t}\right] \exp\left[ie\frac{\vec{r}'-\vec{r}}{t'-t} \int_t^{t'} \vec{A}(\tau)d\tau\right] \times \exp\left\{-i\frac{e^2}{2m} \left[\int_t^{t'} \vec{A}^2(\tau)d\tau - \frac{1}{t'-t} \left[\int_t^{t'} \vec{A}(\tau)d\tau\right]^2\right]\right\}.$$
(2.12)

The  $\vec{A}^2$  term in Eq. (2.11) contributes to an effective mass (see Sec. III). The essential field dependence of the total rate  $\Gamma$  is concentrated in the factors

$$\exp\{ie[\vec{A}(t')\vec{r}' - \vec{A}(t)\vec{r}']\}$$

and the Volkov Green's function  $G_{(e)}$ . In Ref. 1 the entire effect of changing the degree of forbiddenness of the nuclear decay is derived from the factors  $\exp(ie\vec{A}\vec{r})$  in Eq. (2.4). We shall now show explicitly that these factors actually cancel out of the total decay rate against corresponding terms in  $G_{(e)}$  and hence cannot give rise to the effects calculated in Ref. 1.

By inserting Eqs. (2.12) and (2.10) into the total decay rate (2.9), we see that in the field-free limit the major contribution to the time integral in Eq. (2.9) comes from time differences t'-t, for which the phase of the integrand is stationary:

$$|t'-t| = \left[\frac{m(\bar{\tau}' - \bar{\tau}')^2}{2(E_0 - E_v)}\right]^{1/2} . \tag{2.13}$$

The estimate (2.13) is not significantly affected by the external field, in particular, when there is no energy transfer from the field to the electron, as Reiss assumes.

It was shown in Ref. 7 that in the classical limit  $\hbar \to 0$  only times t' infinitesimally close to t contribute to the total transition rate  $\Gamma$ . In this limit the field-dependent exponentials in the integrand of Eq. (2.9) are unity and only the field-independent part survives. Thus in the classical limit the total decay rate would be unaffected by the external field. This is in agreement with the results of Ref. 7.

The remaining field dependence in  $\Gamma$  is then only due to quantum effects. It will be found to be very small for the parameters of Ref. 1, as we shall now demonstrate. We can rewrite the space- and field-dependent exponential in the electron Green's function (2.12) with the help of integration by parts and find

$$\exp\left[ie\frac{\vec{\tau}' - \vec{r}}{t' - t}\int_{t}^{t'}d\tau\,\vec{A}(\tau)\right] = \exp\left[ie\left[\vec{A}(t')\,\vec{\tau}' - \vec{A}(t)\,\vec{\tau}\right]\right] \exp\left[ie\int_{t}^{t'}d\tau\,\vec{R}(\tau)\vec{E}(\tau)\right],\tag{2.14}$$

with

$$\vec{R}(\tau) = \vec{r}' - \frac{\vec{r}'' - \vec{r}}{t' - t}(t' - \tau) \quad . \tag{2.15}$$

By inserting Eq. (2.12) with Eq. (2.14) into Eq. (2.9), we realize that the phase factors  $\exp(ie\vec{A}\vec{r})$  cancel in the total decay rate and we obtain

$$\Gamma = \frac{1}{(2\pi)^6} \lim_{\vec{t} \to \infty} \frac{1}{T} \int d^3r \, d^3r' \int dt \, dt' \exp[iE_0(t'-t)] G_{(r)}(\vec{\tau}',t';\vec{\tau},t)$$

$$\times \left( \frac{2m\pi}{i(t'-t)} \right)^{3/2} \exp\left[i\frac{m}{2} \frac{(\vec{\tau}'-\vec{\tau})^2}{t'-t} \right] \exp\left[ie \int_t^{t'} d\tau \, \vec{R}(\tau) \vec{E}(\tau)\right]$$

$$\times \exp\left\{-i\frac{e^2}{2m} \left[ \int_t^{t'} d\tau \, \vec{A}^2(\tau) - \frac{1}{t'-t} \left\{ \int_t^{t'} d\tau \, \vec{A}(\tau) \right\}^2 \right] \right\}$$

$$\times \left[ \phi_f^*(\vec{\tau}) (gV) \phi_f(\vec{\tau}) \right] \left[ \phi_f^*(\vec{\tau}') (gV)^* \phi_f(\vec{\tau}') \right]. \tag{2.16}$$

In Ref. 1, Eq. (74), the nuclear intensity parameter z,

$$z = (e|\vec{A}|R_0)^2$$
, (2.17)

which specifies the magnitude of the phase  $\exp(i\vec{e} \vec{A} \vec{r})$ , was assumed to be of order of unity. The actual remaining exponential with a field and space dependence in Eq. (2.16) contains the integral

$$\left| e \int_{t}^{t'} \vec{R}(\tau) \vec{E}(\tau) d\tau \right| \sim eR_{0} |\vec{E}| |t' - t|$$

$$\sim eR_{0} \omega |\vec{A}| \left( \frac{mR_{0}^{2}}{E_{0}} \right)^{1/2} \sim \frac{1}{80} \frac{\omega}{E_{0}} << 1.$$
(2.18)

Here  $R_0$  denotes the nuclear radius, and the length of the time interval was estimated by Eq. (2.13). We furthermore determined the amplitude of the electric field by  $|\vec{E}| = \omega |\vec{A}|$ , corresponding to a monochromatic plane wave with frequency  $\omega$ , and applied Eq. (82) of Ref. 1. Equation (2.18) shows that the field- and space-dependent exponential in Eq. (2.16) is unity to an excellent approximation, i.e., the factors  $\exp(ie\vec{A}\vec{r})$ , which are the origin of the large enhancement obtained in Refs. 1 and 2, cancel. The  $\vec{A}^2$ 

terms in Eq. (2.16) do not depend on  $\vec{r}$  and can therefore not change the degree of forbiddenness of the decay.

We also see from the estimate (2.18) that the actual parameter that governs the field impact on the nuclear lifetime is

$$z' = (e\omega |\vec{A}|R_0^2)^2 = z(\omega R_0)^2$$
, (2.19)

which is much smaller than z. This means that one needs a much higher field intensity, or fields with a much shorter wavelength than the one applied in Refs. 1 and 2, to produce a noticeable effect of the external field on the nuclear lifetime.

# III. RELATIVISTIC THEORY

We shall now address ourselves to a relativistic treatment of the electron in analogy with Ref. 1. For a quantitative analysis this is indispensable because the electron energy will, in general, be relativistic, and because the entire decay process is intrinsically relativistic. However, the crucial point of the preceding analysis, the actual replacement of the superficial appearing gauge factors  $\exp(ie\vec{A}\vec{r})$  by  $\exp[ie\int d\tau \vec{R}(\tau)\vec{E}(\tau)]$  in the total decay rate, will proceed

completely analogously in the relativistic treatment. We then shall have to examine the remaining expressions for possible additional terms incorporating  $\vec{A} \vec{r}$  which might yield the large enhancements claimed by Reiss. These will have to be relativistic quantum terms induced by a radio frequency field. Hence it is not likely that they will play any significant role, and, in fact, they will not turn out to do so.

The starting point will now be the relativistic S-matrix element  $S_{fi}$  as given in Eq. (7) of Ref. 1. As we did in Eq. (2.8), we shall again sum over the electron's final momenta p and spins s, obtaining

$$R = \int \frac{d^3 \overline{p}}{2p_0} \sum_{\text{spins}} |S_{fi}|^2$$

$$= \int d^4 p \, \theta(p_0) \delta(p^2 - m^2) \, \sum |S_{fi}|^2$$

$$= \frac{G^2}{4m} \int d^4 x \, d^4 x' [\overline{\Psi}_f(x) \gamma_\mu (1 - \kappa \gamma_5) \Psi_i(x)]$$

$$\times [\overline{\Psi}_i(x') (1 + \kappa \gamma_5) \gamma_\rho \Psi_f(x')]$$

$$\times [\overline{\Psi}_{(\nu)}(x') (1 + \gamma_5) \gamma^0 G^{(+)}(x', x)$$

$$\times \gamma^\mu (1 - \gamma_5) \Psi_{(\nu)}(x)] . \tag{3.1}$$

Here

$$G^{(+)}(x',x) = \int d^4p \; \theta(p_0) \delta(p^2 - m^2) E(x',p) (\not p + m) \overline{E}(x,p)$$
(3.2)

is the homogeneous positive frequency Volkov Green's function replacing Eq. (2.11), and we have written the Volkov solution in the form

$$\Psi_{(e)}(x,p) = E(x,p)u(p,s) ,$$

where u(p,s) is the free Dirac spinor so that

$$(\mathbf{F} - m)u(\mathbf{p}.\mathbf{s}) = 0.$$

We are again using the Coulomb gauge so that the nuclear wave functions are given by Eq. (2.4).

The quantity R, when summed over the final states of the nucleus and the neutrino, yields the total decay rate of Eq.

(2.8). The decisive point is that any Green's function in the presence of an external field  $A^{\mu}(x)$  can be split as

$$G^{()}(x',x) = \phi(x',x)\tilde{G}^{()}(x',x)$$
, (3.3)

where

$$\phi(x',x) = \exp\left[-ie\int_{-x'}^{x'} d\overline{x}_{\mu} A^{\mu}(\overline{x})\right]$$
 (3.4)

is Schwinger's line integral which is to be integrated over a straight line connecting x with x' [the definition can be given in a path-independent way (see Ref. 8)], and  $\tilde{G}^{(\cdot)}(x',x)$  is gauge invariant, i.e., depends only on  $\tilde{E}$  and  $\tilde{B}$ . Hence the entire gauge dependence is isolated in the line integral (3.4). Collecting now all the  $\tilde{r}$   $\tilde{A}$ -dependent exponentials in Eq. (3.1) we obtain

$$\exp\left\{ie\left[\vec{r}\vec{A}(t) - \vec{r}'\vec{A}(t')\right] - ie\int_{x}^{x'}d\vec{x}_{\mu}A^{\mu}(\vec{x})\right\}.$$
(3.5)

Evaluating the line integral yields

$$\int_{x}^{x'} d\overline{x}_{\mu} A^{\mu}(\overline{x}) = -\frac{\overline{x}' - \overline{x}}{k(x' - x)} \int_{kx}^{kx'} d(k\overline{x}) \overline{A}(k\overline{x}) ,$$

where  $k_{\mu}$  denotes the wave vector of the field  $A_{\mu}(kx)$ . In the long wavelength approximation, which is finally also adopted in the relativistic treatment of Ref. 1, Eq. (3.6) reproduces the expression which we already encountered in the nonrelativistic treatment. The exponential (3.5) is then identical to the corresponding exponential in Eq. (2.9) with the nonrelativistic Green's function (2.12), and the arguments pointed out, following Eq. (2.12), apply to the relativistic case as well.

Finally, in order to make sure that the gauge-invariant remainder  $\tilde{G}^{(+)}$  in Eq. (3.3) does not, so to speak by the back door, reintroduce corresponding exponentials, we shall now write down the complete Green's function  $G^{(+)}(x',x)$ . The easiest way to obtain it is by straightforward evaluation of Eq. (3.2), given the Volkov wave functions E(x,p). The analogous approach has been followed, e.g., in Ref. 9, in the case of the Feynman propagator. The result (for arbitrary polarization, i.e.,  $A^{\mu} = \sum_{i=1}^{2} e_i^{\mu} A_i(\xi)$ ,  $\xi = kx$ ,  $ke_i = 0$ ,  $e_i e_j = -\delta_{ij}$ ) is very closely related to the former and reads

$$\tilde{G}^{(+)}(x',x) = \int_{-\infty}^{\infty} \frac{ds}{s^{2}} \exp\left[-is(m^{2}+T) - i\frac{(x-x')^{2}}{4s}\right] \\
\times \left\{\theta[s(\xi'-\xi)]\left[\frac{1}{2s}(x'-x) + m - 2msk\xi_{i}M_{i} - \ell_{i}(\xi'-\xi)L_{i} + i(\xi'-\xi)\gamma_{s}\ell_{i}\epsilon_{ij}M_{j} + k\left[-(x'^{i}-x')L_{i} + s(\xi'-\xi)(L_{i}^{2}-M_{i}^{2}) - \frac{sT}{\xi'-\xi}\right] + i\gamma_{s}k'[-\epsilon_{ij}(x'^{i}-x')M_{j} + 2s(\xi'-\xi)\epsilon_{ij}L_{i}M_{j}]\right] + i\operatorname{sgn}(s)k'\delta(\xi'-\xi)\right\}.$$
(3.7)

Here  $x^i$  denotes the two vector components of  $x^{\mu}$  transverse to  $k^{\mu}$ . We adopt a summation convention for the indices i and j extending from 1 to 2, and  $\epsilon_{ij} = -\epsilon_{ji}$ ,  $\epsilon_{12} = 1$ . The functions L, M, and T are given by

$$T(\xi',\xi) = \frac{e^2}{\xi'-\xi} \int_{\xi}^{\xi'} d\eta A_i(\eta) \left[ A_i(\eta) - \frac{1}{\xi'-\xi} \int_{\xi}^{\xi'} d\eta' A_i(\eta') \right] , \qquad (3.8)$$

$$M_i(\xi',\xi) = -\frac{e}{2(\xi'-\xi)} [A_i(\xi') - A_i(\xi)] , \qquad (3.9)$$

$$L_i(\xi',\xi) = \frac{e}{2(\xi'-\xi)} \left[ A_i(\xi') A_i(\xi) - \frac{2}{\xi'-\xi} \int_{\xi}^{\xi'} d\eta A_i(\eta) \right] . \tag{3.10}$$

We incidentally note that the quantity  $T(\xi', \xi)$  in view of Eq. (3.7) specifies a space-time-dependent mass correction. It already showed up in the nonrelativistic treatment [cf. the last exponential in Eq. (2.12)].

We now have to check whether Eq. (3.7) contains any terms similar to  $\cos(e\vec{a}\vec{r} - e\vec{a}\vec{r}')$ , which is the sole cause of the enhancement in Ref. 1 [cf. Eq. (110)]. There are two candidates in Eq. (3.7):

$$-K(x''-x')L_i$$
,  $-i\gamma sK\epsilon_{ij}(x''-x')M_j$ .

We notice that they are both only linear in (x''-x'). Hence they can at best reduce the order of forbiddenness by one. Since they are proportional to K, their order of magnitude is  $\omega e |\vec{A}| R_0 = \omega \sqrt{z}$ , which has to be compared with the electron mass m in the square bracket in Eq. (3.7). Since  $\omega/m << 1$ , in particular, for a radio frequency field, whatever effect is caused by these terms will be very small, as was to be expected.

# IV. AN ALGEBRAICAL ERROR IN REF. 1

The above considerations strongly suggest that there is a computational error in Ref. 1. Hence we have cursorily checked some of the calculations. In fact, it appears that factors  $(-i)^j$  and  $i^m$  are missing on the right-hand side of Eqs. (56) and (57) of Ref. 1, respectively, which should

$$\exp(-ie\vec{A}\vec{r}) = \exp[-ie\vec{a}\vec{r}\cos(kx+\rho)]$$

$$= \sum_{j=-\infty}^{\infty} J_j(e\vec{a}\vec{r})\exp[-ij(kx+\rho)](-i)^j$$
(4.1a)

and

$$\exp(ie\vec{A}'\vec{\Gamma}') = \exp[ie\vec{a}\vec{\Gamma}'\cos(kx'+\rho)]$$

$$= \sum_{m=-\infty}^{\infty} J_m(e\vec{a}\vec{\Gamma}') \exp[im(kx'+\rho)]i^m . (4.1b)$$

These factors seem to be consistently missing. If they are included, the crucial equation (110) of Ref. 1,

$$\sum_{q} (-)^{q} J_{1q}(e \, \overline{a} \, \overline{r} - e \, \overline{a} \, \overline{r}') = \cos(e \, \overline{a} \, \overline{r} - e \, \overline{a} \, \overline{r}') \quad , \quad (4.2)$$

loses the factor  $(-)^{\P}$  on the left-hand side and is changed into

$$\sum_{i} J_{2q}(e\vec{a}\vec{r} - e\vec{a}\vec{r}') = 1 , \qquad (4.3)$$

and there is no field induced enhancement of forbidden beta decay, in agreement with the arguments previously put forward in this paper.

# V. REVIEW AND CRITICISM OF THE MTA WAVE FUNCTION

For a review of the MTA wave function from Reiss's point of view we refer to Ref. 4. The MTA method claims to be "gauge specific" for the Coulomb gauge (C gauge; vector potential  $\vec{A}$  and vanishing scalar potential  $A_0 = 0$ ). The Schrödinger equation in the long-wavelength approximation for the vector potential  $\vec{A}$  then has the form

$$i\frac{\partial}{\partial t}\Psi(\vec{r},t) = [H_0 + H_1(t)]\Psi(\vec{r},t) , \qquad (5.1)$$

with

$$H_0 = \frac{1}{2m} \vec{p}^2 + V(\vec{r}), \quad H_1(t) = -\frac{e}{m} \vec{A}(t) \vec{p} + \frac{e^2}{2m} \vec{A}^2(t)$$
 (5.2)

If one writes an ansatz for  $\Psi(\vec{r},t)$  in the form

$$\Psi(\vec{r},t) = \exp[ie\vec{A}(t)\vec{r}]\Phi(\vec{r},t) , \qquad (5.3)$$

then the new wave function  $\Phi(\vec{r},t)$  obeys the equation of motion

$$i\frac{\partial}{\partial t}\Phi(\vec{r},t) = [H_0 + H_2(t)]\Phi(\vec{r},t) , \qquad (5.4)$$

with

$$H_2(t) = -e\vec{E}(t)\vec{r} . \qquad (5.5)$$

The so-called "momentum translation approximation" consists of neglecting the perturbation  $H_2$  in Eq. (5.4) and replacing  $\Phi$  in Eq. (5.3) by the unperturbed wave function  $\Phi_0$ , given by

$$i\frac{\partial}{\partial t}\Phi_0(\vec{r},t) = H_0\Phi_0(\vec{r},t) . \qquad (5.6)$$

We then obtain the MTA wave function as an approximation of the exact solution  $\Psi$ :

$$\Psi_0(\vec{r},t) = \exp[ie\vec{A}(t)\vec{r}]\Phi_0(\vec{r},t) . \qquad (5.7)$$

This procedure is considered to be justified if  $H_2$  is much smaller than  $H_1$ , in particular, when  $H_1$  is too large to be treated as a perturbation with respect to  $H_0$ , whereas  $H_2$  still is a small perturbation compared with  $H_0$  (i.e., the magnitude of  $H_2$  is small as compared with a characteristic energy of the field-free problem). The condition " $H_2$  small compared with  $H_1$ " means that the transition matrix elements from an initial unperturbed state |f| to a final unperturbed state |f| are much smaller when the transition is induced by the residual interaction  $H_2$  instead of  $H_1$ . The states |f|

and  $|f\rangle$  are eigenstates of  $H_0$ :

$$H_0(n) = E_n(n) \quad . \tag{5.8}$$

By using the commutator relation

$$[\vec{r}.H_0] = i\frac{\vec{p}}{m} . \tag{5.9}$$

as well as

$$|\vec{\mathbf{E}}| = \omega |\vec{\mathbf{A}}| \qquad (5.10)$$

we find

$$\frac{|\langle f|H_2(t)|i\rangle|}{|\langle f|H_1(t)|i\rangle|} \sim \frac{\omega}{E_i - E_f}$$
 (5.11)

Hence in situations where the field frequency is much smaller than the considered transition energy, the MTA procedure might seem to be well justified.

As we have noted, central to the MTA is the condition that the transition matrix elements for the interaction  $H_2$  are much smaller than for its counterpart  $H_1$ . This applies, in particular, to nuclear transitions in the presence of an optical or even radio frequency field. But one must be careful in drawing conclusions from or making approximations based upon the estimate (5.11). Instead of transition matrix elements, one actually has to compare transition probabilities, which are the directly measurable quantities. As we shall see below, the factor  $\omega/(E_i - E_f)$  does not occur after integration over time and the two interactions  $H_1$  and  $H_2$  give the same transition probabilities for any ratio of  $\omega$  over  $(E_i - E_f)$ .

Let us repeat a supposedly well known argument. In the interaction picture the transition amplitude to first order of perturbation theory reads

$$F(T) = -i \int_0^T dt \exp[-i(E_i - E_f)t] \langle f | H_{\text{int}}(t) | i \rangle .$$
(5.12)

If we use the interaction  $H_{int} = H_2$ , we find

$$F(T) = ie \langle f | \vec{\tau} | i \rangle \int_0^T dt \exp[-i(E_i - E_f)t] \vec{E}(t) . \quad (5.13)$$

For the case  $H_{\text{int}} = H_1$ , let us consider the situation where the field is switched on at time t = 0 and that we are only interested in the transition probability at time T, when the field is already switched of f, so that

$$\vec{A}(0) = 0, \quad \vec{A}(T) = 0$$
 (5.14)

This is the usual experimental situation. We can then rewrite the integral in Eq. (5.12) by partial integration

$$\int_0^T \exp[-i(E_i - E_f)t] \vec{A}(t) dt$$

$$= \frac{i}{E_i - E_f} \int_0^T \exp[-i(E_i - E_f)t] \vec{E}(t) dt . \quad (5.15)$$

We have now obtained a factor  $(E_1 - E_f)$  by replacing  $\vec{A}$  by  $\vec{E}$  in the integrand, instead of the factor  $\omega$ , which entered the relation (5.11) via Eq. (5.10). By using Eqs. (5.9) and (5.15) we find for  $H_{int} = H_1$  again the transition amplitude (5.13).

This simple calculation shows that the transition probability is the same whether one uses the interaction  $H_1$  or  $H_2$ .

This holds true to any order in the perturbation series. We can furthermore drop the restriction (5.14) and obtain equal transition amplitudes F(T) at any time T, if we take into account that the same physical state is represented by different state vectors in different gauges if  $\overline{A}(0) \neq 0$  or  $\overline{A}(T) \neq 0$  (see next paragraphs). The error will therefore be of the same order of magnitude whether one neglects the interaction  $H_2$  in Eq. (5.4) or  $H_1$  in Eq. (5.1) for any ratio of the field frequency  $\omega$  over the typical transition energy  $E_1 - E_f$ . Since the approximate solution  $\Phi_0$  of Eq. (5.4) is an unperturbed wave function, these arguments indicate that also the MTA wave function represents a noninteracting state.

In order to see that the MTA wave function represents nothing but a noninteracting state, we must consider the different gauges involved in the problem. The wave functions of all charged particles participating in any reaction or decay process must be taken consistently in the same gauge. The Coulomb gauge is convenient for the calculation of the Volkov wave function of a free particle in the presence of an external electromagnetic field. To derive the properties of the nonrelativistic MTA wave function it is more instructive to begin with the electric field gauge [E] gauge; vanishing vector potential  $\vec{A} = 0$  and scalar potential  $-e\vec{E}(t)\vec{r}$ . Wave functions are transformed from the C gauge (notation  $\Psi$ ) to the E gauge (notation  $\Phi$ ) by the unitary transformation

$$\Phi(\vec{r},t) = \exp[-ie\vec{A}(t)\vec{r}]\Psi(\vec{r},t) , \qquad (5.16)$$

where  $\vec{A}$  is the vector potential in long wavelength approximation for the C gauge.

The Schrödinger equation in the E gauge has the form (5.4), which was derived at the beginning of this chapter in a different context. If we entirely neglect the interaction of the particle with the field to a zeroth approximation, we obtain from Eq. (5.6) as an approximate solution the wave function  $\Phi_0$  in the absence of the external field.

In the present example of beta decay, the nuclear state has to be determined from the Schrödinger equation in the C gauge, i.e., from Eq. (5.1), since the electron wave function is given in the C gauge. If we approximate the solution of Eq. (5.1) by the noninteracting state  $\Phi_0$  in the E gauge and use the gauge transformation (5.16), we obtain the wave function  $\Psi_0$  of Eq. (5.7), the MTA wave function. Although  $\Psi_0$  depends on the vector potential  $\vec{A}$ , it still represents a noninteracting state since a state vector which does not include any interaction with the external field in a particular gauge (here the E gauge), neither does so when transformed to any other gauge (here the C gauge).

It should be mentioned that the wave function  $\Phi_0$  is not the correct noninteracting solution in the C gauge. In the C gauge the operator of the canonical momentum  $\vec{p} = -i \vec{\nabla}$  differs from the operator of the kinetical momentum. Hence in the C gauge the Hamiltonian  $H_0$  in Eq. (5.2) does not describe the situation, where the interaction of the particle with the field is entirely neglected. The correct procedure is first to transform from the C gauge via (5.16) to the E gauge, where the vector potential vanishes and the operators of the kinetical and canonical momentum are identical, so that  $p^2/2m$  is the operator of the kinetical energy. Hence in this gauge the Hamiltonian in Eq. (5.6) really specifies the field-free motion, and  $\Phi_0$  represents the state in the absence of the field. The wave function in the C

gauge, which completely neglects the interaction with the field, is then given by Eq. (5.7), i.e., by the MTA wave function. For a lucid and thorough discussion of gauge invariance in quantum mechanics see Ref. 5.

Applying this discussion to the nuclear beta decay, we see that the S matrix (2.1) includes the exact electron state and noninteracting nuclear states in the C gauge. In the nonrelativistic approach the S matrix could also be written in the E gauge. The unperturbed nuclear state is then given by the field-free wave function  $\Phi_0$ , and the gauge phase  $\exp(ie\vec{A}\vec{r})$  is incorporated in the corresponding Volkov wave function, which we obtain from Eq. (2.3) and the transformation (5.16). The entire field dependence is then concentrated in the electronic wave function. This procedure yields the same analytical form of the S-matrix element as Eq. (2.1).

The factor  $\exp(ie \vec{A} \vec{r})$  always appears when one consistently combines the nuclear state in the absence of the field, which is related to the E gauge, with a Volkov state, which can be calculated in a simple way for the C gauge. This factor can be shifted from the nuclear to the electronic wave function in different gauges and has nothing to do with the interaction of the nucleus with the field. The identification of the factor  $\exp(ie\vec{A}\vec{r})$  in Eq. (5.3) as a gauge transformation was in the present context first pointed out in Ref. 6. In Ref. 4, Reiss does not consider this factor as a gauge transformation but rather calls it a "unitary transformation within the Coulomb gauge." He then claims that due to this phase factor the MTA wave function fairly represents the effects of the applied field on the particle to any order of interaction. Relying on this interpretation, Reiss attempts in Ref. 1 to derive the entire effect of changing the degree of forbiddenness of a nuclear decay and of enhancing nuclear decay rates from this factor  $\exp(ie \vec{A} \vec{r})$ .

If the interaction of the nucleus with the field is neglected, only the coupling of the electron with the field can still modify the nuclear lifetime. But as it was shown in Ref. 7, this can only happen for a field which is much stronger than the one considered in Ref. 1, as long as the field frequency is very small as compared with the nuclear decay energy. Therefore the model used in Ref. 1 cannot result in an appreciable change of the nuclear lifetime.

# V. SUMMARY

The basic formalism developed at length in Ref. 1 is essentially the same as in Refs. 12 and 13 (cf. Ref. 14). The theory of Ref. 1 only differs from this previous work by using the correct noninteracting wave function  $\Psi_0$  (2.4) instead of  $\Phi_0$  (2.5) (but with the misguided intention of incorporating the interaction with the field) and by considering linear polarization of the field instead of circular polarization. Whether the field is linearly or circularly polarized is rather immaterial for the effects in question. Considering linear in place of circular polarization mainly increases the calculational labor without adding additional insight.

We would like to concentrate on two central objections to Refs. 1 and 2: (i) Contrary to its intentions, Reiss's model

does not include any interaction between the nucleus and the field. Notably, the so-called momentum-translation (MTA) wave function is nothing but the correct free wave function in the Coulomb gauge, as was shown in Sec. V in three different ways. (ii) The total decay rate cannot be influenced by an external field as chosen in Ref. 1, if one describes the electron by the relativistic Volkov wave function and the nucleus by the nonrelativistic MTA wave function. This can be derived by general arguments from point (i) and Ref. 7. We also prove this explicitly in Secs. II and III. There we show that in a correct treatment the gauge factors  $\exp(ie\vec{A}\vec{r})$  (from which Ref. 1 derives its entire effect) mutually cancel in the total decay rate. Our argument shows that this cancellation is independent of the polarization and pulse shape of the external field. The dramatic enhancements of Refs. 1 and 2 seem to be due to an algebraic error. When this is corrected no enhancement remains, in agreement with the results of Secs. II and III.

The physical concept underlying Refs. 1 and 2 differs from the previous work (Refs. 12 and 13) by intending to concentrate on forbidden beta decay in the presence of an intense radio frequency field rather than an optical field. Some final remarks on these two new aspects: (a) Changing the degree of forbiddenness of a nuclear decay by modifying the nuclear states under the impact of an external field is an exciting idea, but cannot be achieved by Reiss's model, which only uses noninteracting nuclear states. In particular, it should be emphasized that Reiss tries to treat forbidden beta decay along the same lines as allowed beta decay, i.e., a multipole expansion of the lepton wave functions and relativistic corrections to the nuclear wave functions, both of which normally enable forbidden decays to take place, are not considered, since the dominant mechanism of the enhancement is supposed to originate from the gauge factors  $\exp(ie A \vec{r})$ . (b) The advantage of radio frequencies as compared with optical frequencies seems to lie in the fact that larger values of the parameter  $(ea/m)^2$  can be achieved at much lower field strengths. But if very high intensity radio frequency fields with a wavelength of  $\lambda \sim 100$  m (Ref. 2) are applied, the applicability of the Volkov solution, which assumes a plane wave field of infinite extent in space and time, seems very doubtful and certainly requires some justification. A relativistic electron which moves freely in such a field performs an oscillatory motion over a distance of  $\lambda$ . Such electromagnetic fields also raise experimental problems, since the atomic electrons tend to shield an external low frequency field, reducing its field strength at the site of the nucleus by orders of magnitude. 15

We would finally like to emphasize that it remains an open question as to whether properly including the interaction between the nucleus and the field might yet lead to some enhancement of forbidden beta decay, although we believe that the orders of magnitude stated in Refs. 1 and 2 are very unlikely to be achieved by the latter mechanism.

We benefited from discussions with D. H. Kobe and P. Zoller. This work was supported in part by the Air Force Office of Scientific Research under Contract No. AFOSR-81-0128.

- <sup>1</sup>H. R. Reiss, Phys. Rev. C <u>27</u>, 1199 (1983).
- <sup>2</sup>H. R. Reiss, Phys. Rev. C 27, 1229 (1983).
- <sup>3</sup>D. M. Volkov, Z. Phys. 94, 250 (1935).
- <sup>4</sup>H. R. Reiss, Phys. Rev. A 23, 3019 (1981).
- <sup>5</sup>K.-H. Yang, Ann. Phys. (N.Y.) 101, 62 (1976).
- 6C. Cohen-Tannoudji, J. Dupont-Roc, C. Fabre, and G. Grynberg, Phys. Rev. A 8, 2747 (1973).
- <sup>7</sup>W. Becker, G. T. Moore, R. R. Schlicher, and M. O. Scully, Phys. Lett. <u>94A</u>, 131 (1983).
- <sup>8</sup>J. Schwinger, Phys. Rev. 82, 664 (1951); Schwinger's derivation is carried out for the Feynman propagator  $G^{(F)}$ ; however, one can easily convince oneself that the factorization (3.3) applies to any Green's function.
- <sup>9</sup>H. Mitter, Acta Phys. Austriaca Suppl. XIV, 397 (1975).
- <sup>10</sup>H. R. Reiss, Phys. Rev. A 1, 803 (1970).
- 11 J. Forney, A. Quattropani, and F. Bassani, Nuovo Cimento

- **B37**, 78 (1977).
- <sup>12</sup>i G. Baranov, Izv. Vyssh. Uchebin Zaved. Fiz. 4, 115 (1974) [Sov. Phys. J. 4, 533 (1975)].
- <sup>13</sup>W. Becker, W. H. Louisell, J. D. McCullen, and M. O. Scully, Phys. Rev. Lett. <u>47</u>, 1262 (1981).
- 14Whereas the basic formalism developed in Refs. 12 and 13 is—essentially correct, the quantitative estimates are not. The predicted enhancements of allowed beta decay are nonexistent for the fields considered there and are due to a too careless evaluation of sums over Bessel functions. This has been corrected in Ref. 7, where the physical reason for the fact that total decay rates are virtually unaffected by external plane wave fields is pointed out.
- 15J. I. Gersten and M. H. Mittleman, Phys. Rev. Lett. 48, 651 (1982).

# END

# FILMED

1-85

DTIC